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THE COMBINED ELLIPSOMETRIC METHOD OF COMPLETE OPTICAL CHARACTERIZATION OF CRYSTALS.
III. EXPERIMENTAL DETERMINATION OF THE ORIENTATION OF OPTICAL AXIS IN CRYSTAL

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In the third part of the article, devoted to the description of the combined ellipsometric method for the complete optical characterization of crystals, two variants of the technique for experimentally determining the orientation of optical axes using ellipsometry are presented. The first variant is based on the search for the circular cross section of the optical indicatrix. As is known, this cross section is perpendicular to the optical axis of the crystal. This is done by successive measurements of the dependence of the effective refractive index on the angle of rotation of the crystal around the normal to the plane under study, $n_{eff} = f(\alpha)$. The second variant is based on the fact that the effective refractive index, n_{eff}, does not depend on the angle of incidence φ of the beam on the plane under study, if the angle between the incident beam and the optical axis does not change. Thus, if measurements are performed on the plane of the optical axes of the crystal, then the problem is reduced to finding a plane that is perpendicular to the optical axis. This is done by successive measurements of the dependence of the effective refractive index on the angle of incidence of the beam on the plane of the optical axes, $n_{eff} = f(\varphi)$. The second variant of this technique was tested by ellipsometric measurements of a $CdWO_4$ crystal made on the plane of the optical axes. It has been established that the angle between the optical axes (the bisector of the angle is the principal axis $\boldsymbol{N_g}$ of the optical indicatrix) is equal to $2V = 92° \pm 1°$. Since $2V > 90°$, the $CdWO_4$ crystal is an optically negative crystal (the optical sign is minus).

Key words: ellipsometry, optical indicatrix, principal refractive indexes, uniaxial and biaxial crystals.

1. Introduction

This is the third part of the article devoted to the description of the combined ellipsometric method for the complete optical characterization of crystals. In the first part of this article [1], a method for determining the orientation of an optical indicatrix in crystals was described in detail. This method became the basis of the combined ellipsometric method and its first stage. In the second part of the article [2], we showed how to determine the optical constants of a particular crystal, knowing the orientation of the optical indicatrix and using the relationships obtained by R. H. W. Graves [3]. Analyzing the results of determining the optical constants of an optically biaxial $CdWO_4$ crystal, it is easy to see that the accuracy of determining the orientation of the optical axes in this crystal is not high enough [2]. If we average over the results of measurements in two configurations (see Table 4 [2]), then we obtain the following value of the angle between the optical axes: $2V = 91°40'\pm 1°40'$. This accuracy in determining the *2V* angle led us to ask an obvious question. Is it possible, within the framework of the ellipsometric method under consideration, to propose a technique for more accurate determination of the orientation of the optical axes? It is this technique in two variants that we present in this article. Obviously, we are talking, first of all, about optically biaxial crystals, since, having determined the orientation of the optical indicatrix in an optically uniaxial crystal we automatically determine the orientation of the optical axis (or vice versa).

2. Technique for experimental determination of the orientation of optical axes in a crystal

2.1. The first variant

Recall that in optically biaxial crystals, both optical axes always lie in the principal section of the $\boldsymbol{N_gON_p}$ optical indicatrix (Fig. 1). As in the second part of this article, we will designate the coordinate system of the optical indicatrix, as well as the optical axes, in bold type. Accordingly, the optical indicatrix has two circular cross sections that are perpendicular to the optical axes $\boldsymbol{O_1}$ and $\boldsymbol{O_2}$. The line of intersection of the circular sections is the mean principal axis $\boldsymbol{N_m}$ of the optical indicatrix. It is perpendicular to the plane of optical axes $\boldsymbol{O_1OO_2}$.

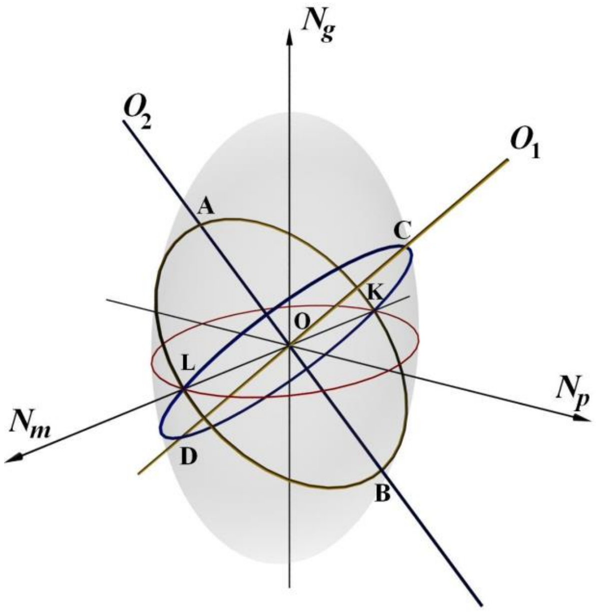

Fig.1. Orientation of the optical axes $\boldsymbol{O_1}$ and $\boldsymbol{O_2}$ relative to the $\boldsymbol{N_mN_pN_g}$ coordinate system of the optical indicatrix in an optically biaxial crystal. Planes AKBL and CKDL correspond to the circular cross sections of the ellipsoid of the optical indicatrix.

The major $\boldsymbol{N_g}$ and minor $\boldsymbol{N_p}$ principal axes of the optical indicatrix are the bisectors of the angles between the optical axes. Usually, the angle *2V* is determined, the bisector of which is the major principal axis $\boldsymbol{N_g}$ of the optical indicatrix. Having determined the values of the principal refractive indices n_g, n_m and n_p (see [2]), the angle V is calculated using the formula [4]:

$$tgV = \sqrt{\frac{n_p^{-2} - n_m^{-2}}{n_m^{-2} - n_g^{-2}}} \tag{1}$$

This is how we determined the angle V in the second part of this article [2] in a $CdWO_4$ crystal. Thus, knowing the value of the angle $2V$, we determine both the orientation of the optical axes in a biaxial crystal and the optical sign of the crystal. However, it should be recalled that optically biaxial crystals are characterized by dispersion both of the principal axes of the optical indicatrix and of the optical axes. Therefore, when speaking about the orientation of the optical indicatrix and optical axes, it is imperative to indicate the wavelength of electromagnetic radiation at which the measurements were made. We have repeatedly pointed out that in our studies all measurements were performed at the He-Ne laser radiation wavelength, λ=632.8 nm.

In the same part of the article, we will consider two variants of direct experimental determination of the orientation of optical axes in an optically biaxial crystal using ellipsometry. On fig. 1 shows the case when the angle $2V$ between the optical axes of the crystal is close to 90°. This is typical, for example, for the $CdWO_4$ crystal, which is our main object for experimental verification of the proposed combined ellipsometric method. In the first variant of this ellipsometric technique for determining the orientation of the optical axes, we use the fact that the dependence $n_{eff} = f(\alpha)$ (see [1]), measured on a plane perpendicular to the optical axis, has the form of a straight line parallel to the abscissa axis. In other words, the effective refractive index, n_{eff}, does not depend on the angle of rotation α of the crystal around the normal to this plane. The same dependence, presented as an analogue of the elliptical cross section of the optical indicatrix (see Fig. 3 [1]), has the form of a circle. Thus, the goal of the first variant of the technique is to find this circular cross section. The method of its search is practically similar to the method of determining the orientation of the optical indicatrix [1]. Knowing the approximate orientation of the optical axes, determined at the second stage of the complete optical characterization of the crystal [2], we prepare for measurements a plane approximately perpendicular to one of the optical axes of the crystal (for example, to the axis $\boldsymbol{O_1}$ (Fig. 2)). In our particular case with a CdWO4 crystal, this will be a plane that is inclined by approximately 45° relative to the principal cross section $\boldsymbol{N_gON_m}$ of the optical indicatrix (Fig. 2). After careful preparation of this plane, we measure the dependence $n_{eff} = f(\alpha)$ on it. Suppose we have obtained an analogue of the elliptic cross section AKBL (Fig. 2). If the preparation of the plane was done correctly, then the values of n_{eff} at points K and L should be the same up to the measurement error. The values of n_{eff} at points A and B must also be the same, but in general they will differ from the values of n_{eff} at points K and L. Let's denote the value of n_{eff} at points A and B as n_{eff1}, and at points K and L as n_{eff0}. If, for example, $n_{eff1} > n_{eff0}$, then we rotate the plane corresponding to the analogue of the elliptical cross section AKBL by approximately 1-1.5° around the mean principal axis $\boldsymbol{N_m}$ of the optical indicatrix in the direction of increasing the angle $2V$.

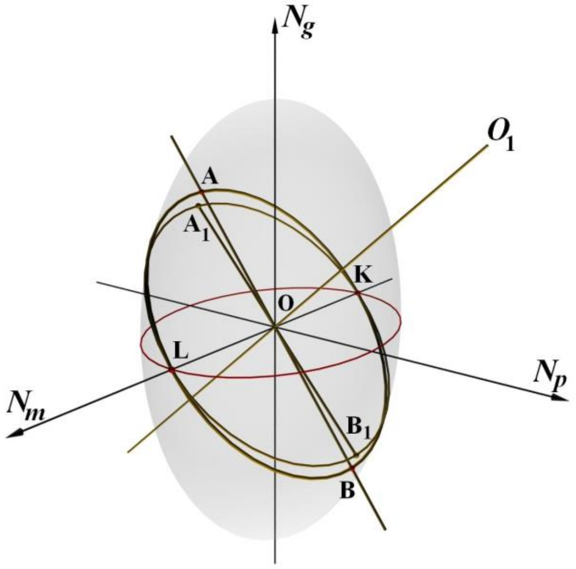

Fig.2. Orientation of two analogues of elliptical cross sections of the optical indicatrix of an optically biaxial crystal relative to the coordinate system of the indicatrix. These analogs were obtained as a result of successive measurements of the dependence $n_{eff} = f(\alpha)$ in order to find the circular cross section of the indicatrix. In this case, the inequality $n_{eff1} > n_{eff2} > n_{eff0}$ holds (see text).

The purpose of this and subsequent similar steps is obvious: to find an analogue of the circular cross section of the optical indicatrix. This rotation is performed by grinding and polishing the new plane. Having prepared this plane in an appropriate way for ellipsometric measurements, we will perform the necessary measurements on it. We will obtain a new dependence $n_{eff} = f(\alpha)$ and, accordingly, a new analogue of the elliptical cross section A_1KB_1L of the optical indicatrix (Fig. 2). Let us emphasize once again: if the orientation of the mean principal axis $\boldsymbol{N_m}$ of the optical indicatrix is determined correctly, then in this case the values of n_{eff} at the points K and L will be the same as for the previous plane. Denoting the value of n_{eff} at points A_1 and B_1 as n_{eff2}, we compare it with n_{eff0}. If it turns out that $n_{eff2} = n_{eff0}$, then the resulting analog of the elliptic section A_1KB_1L is a circle, and thus the goal is achieved. If $n_{eff2} > n_{eff0}$, but $n_{eff2} < n_{eff1}$, then we rotate the A_1KB_1L plane by 1-1.5° in the same direction and again measure the dependence $n_{eff} = f(\alpha)$. In other words, the procedure is completely repeated.

If it turns out that $n_{eff2} < n_{eff0}$, then the plane should be rotated in the opposite direction, but not more than 1°. Having carefully prepared a new plane for measurements, let us measure the dependence $n_{eff} = f(\alpha)$ on it. We will obtain a new analogue of the elliptic cross section A_2KB_2L (Fig. 3). Let us denote the value of the effective refractive index at points A_2 and B_2 as n_{eff3}. Taking into account the accuracy of measurements, achievable, for example, on the LEF-3M-1 ellipsometer (λ=632.8 nm), we can state with confidence that, up to the measurement error, we will obtain an approximate equality, $n_{eff3} \approx n_{eff0}$. Consequently, the cross section A_2KB_2L will be one of the two circular cross sections of the optical indicatrix. Thus, we have found a plane perpendicular to the optical axis and, accordingly, determined the orientation of the axis itself.

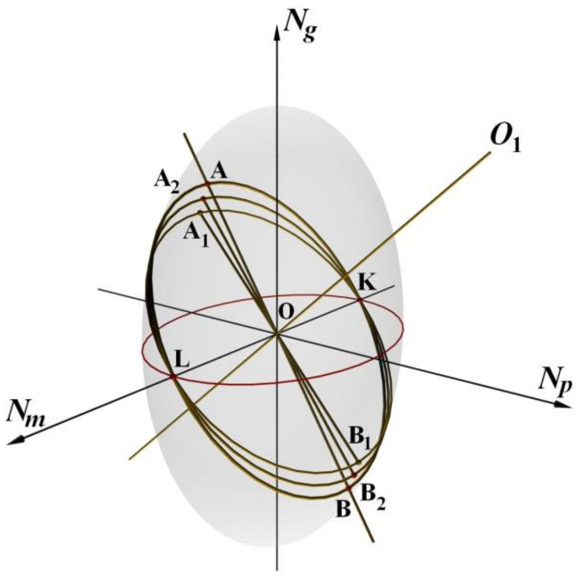

Fig.3. Orientation of three analogues of elliptical cross sections of the optical indicatrix of an optically biaxial crystal relative to the coordinate system of the indicatrix. In this case, the inequality $n_{eff1} > n_{eff0} > n_{eff2}$ and the approximate equality $n_{eff3} \approx n_{eff0}$ hold (see text).

Attention should be paid to one very important experimental fact. The dependence $n_{eff} = f(\alpha)$ can be measured at any angle of incidence φ of the laser beam on the crystal plane under study. It is only important that each specific dependence $n_{eff} = f(\alpha)$ be measured at the same angle of incidence. From this follows an obvious conclusion. The effective refractive index, n_{eff}, does not depend on the angle of rotation of the crystal around the normal to the plane under study, if the angle between the incident beam and the optical axis does not change during rotation. We used the corollary of this conclusion in developing the second variant of the considered experimental technique.

At first glance, it may seem that the considered first variant of the ellipsometric technique for determining the orientation of optical axes is somewhat cumbersome. To this objection we can say the following. First, in laboratories with modern equipment to perform the necessary technological operations for preparing the corresponding crystal planes for measurements, we do not see any difficulties in implementing this variant of the technique. Secondly, taking into account all that has been said, it is obvious that the implementation of this option is possible only in such laboratories.

2.2. The second variant

As we said above, the second variant of the ellipsometric technique for determining the orientation of the optical axes is based on the fact that the effective refractive index, n_{eff}, does not depend on the angle of rotation of the crystal around the normal to the plane under study, if the angle between the incident beam and the optical axis during rotation does not changes. It is clear that the same result should be in case this angle is 90°. Thus, if we measure n_{eff} in the principal cross section $\boldsymbol{N_gON_p}$ of the optical indicatrix, then we can find such a position of the plane of incidence of the beam relative to, for example, the principal axis $\boldsymbol{N_p}$, when this plane is perpendicular to the optical axis. Obviously, with such a mutual orientation of the plane of incidence and the optical axis, the value of n_{eff} should not depend on the angle of incidence φ. This conclusion will be confirmed by the example of the study of the $CdWO_4$ crystal (Fig. 4).

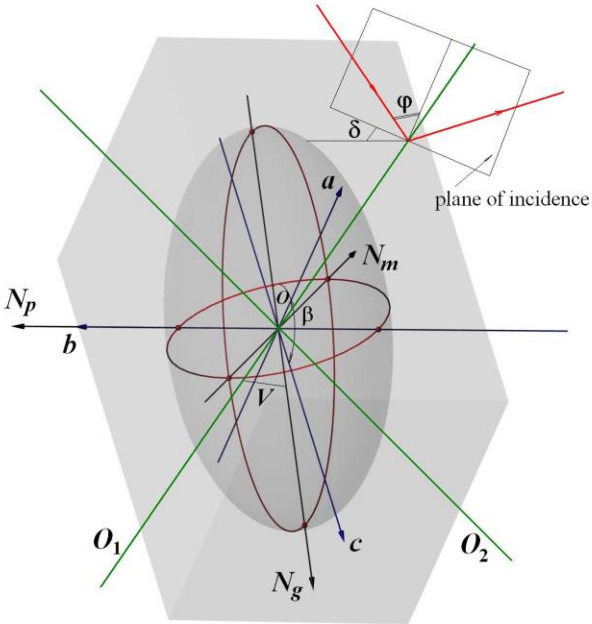

Fig.4. Configuration of ellipsometric measurements of an optically biaxial $CdWO_4$ crystal to determine the orientation of its optical axes. $\boldsymbol{N_mN_pN_g}$ is the coordinate system of the optical indicatrix, ***abc*** is the crystallographic coordinate system. $\boldsymbol{O_1}$ and $\boldsymbol{O_2}$ are the optical axes of the crystal. The measurements are performed on a plane that corresponds to the principal cross section $\boldsymbol{N_gON_p}$ of the optical indicatrix.

Thus, the problem is to find such an angle of rotation δ of the plane of incidence relative to the main axis $\boldsymbol{N_p}$, when the dependence $n_{eff} = f(\varphi)$ will look like a straight line parallel to the abscissa axis. In other words, the effective refractive index, n_{eff}, should not depend on the angle of incidence of the beam on the crystal plane under study. In this case, the perpendicular to the plane of incidence will be one of the two optical axes of the crystal. Accordingly, the required angle V is defined as $V = \delta$. We tested this variant of the technique under consideration by determining the orientation of the optical axes in the $CdWO_4$ crystal (monoclinic syngony, class $2/m$, symmetry space group $P2/c$, a =0.502 nm, b=0.585 nm, c=0.507 nm, β=91.5º [5]). We have studied a $CdWO_4$ crystal alloyed from the melt with PbO (0.375 mass %). The dependence $n_{eff} = f(\varphi)$ was measured on the plane corresponding to the principal cross section $\boldsymbol{N_gON_p}$ of the optical indicatrix (Fig. 4). As is known, in all monoclinic crystals the crystallographic axis ***b*** coincides with one of the principal axes of the optical indicatrix. In the $CdWO_4$ crystal, the ***b*** axis coincides with the principal axis $\boldsymbol{N_p}$. Since the ***b*** axis is perpendicular to the (010) plane, along which the crystal is very easily cleaved, the orientation of this axis (and, respectively, of the axis $\boldsymbol{N_p}$) was determined with high accuracy. It is relative to the principal axis $\boldsymbol{N_p}$ (or, more precisely, relative to the principal cross section $\boldsymbol{N_mON_p}$ of the optical indicatrix) that we oriented the plane of incidence of the beam in the process of measuring the dependence $n_{eff} = f(\varphi)$ (Fig. 4). Figure 5 shows the results of measurements of this dependence, performed at different angles of rotation δ of the plane of incidence relative to the principal cross section $\boldsymbol{N_mON_p}$.

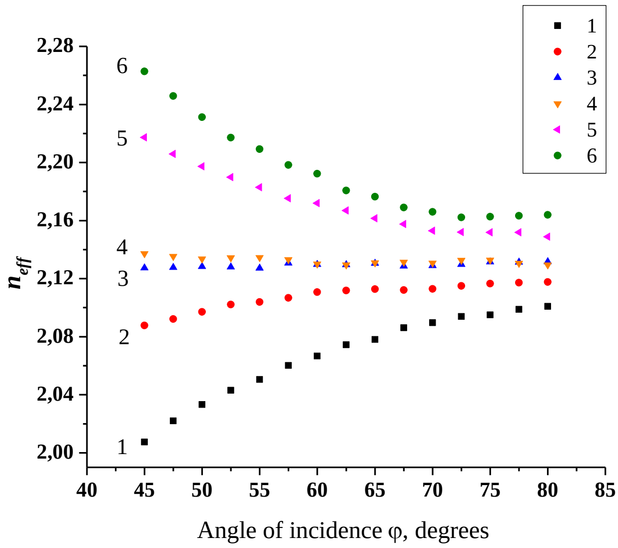

Fig.5. The dependence $n_{eff} = f(\varphi)$ measured on the plane corresponding to the principal cross section $\boldsymbol{N_gON_p}$ of the optical indicatrix of the $CdWO_4$ crystal. The measurements were performed at different angles of rotation δ of the plane of incidence of the laser beam relative to the principal cross section $\boldsymbol{N_mON_p}$: 1 – 0°, 2 – 35°, 3 – 46°, 4 – 48°, 5 – 65° and 6 – 90°.

The analysis showed that the dependence $n_{eff} = f(\varphi)$ is closest to a straight line at δ = 46°±0.5° (curve 3, Fig. 5). Note that this is the maximum possible accuracy achievable in measurements on the LEF-3M-1 ellipsometer, even when studying high-quality crystals. Thus, in this case, the angle $2V = 92° \pm 1°$, and, accordingly, $2V > 90°$. Therefore, a $CdWO_4$ crystal is an optically negative crystal (the optical sign is minus). Summarizing the result obtained, we can confidently state that the $CdWO_4$ crystal is a pronounced optically biaxial crystal with an angle between the optical axes close to 90°. One more important conclusion should be paid attention to, which follows from the results shown in Fig.5. This variant of the technique is most efficient if the dependence $n_{eff} = f(\varphi)$ is measured at small angles of incidence of the laser beam on the plane of the crystal under study. In particular, if we compare curves 3 and 4 (Fig. 5), it is obvious that the results of measurements of the dependence $n_{eff} = f(\varphi)$ at φ > 60° do not allow us to accurately determine the required angle δ. Based on the technical characteristics of the LEF-3M-1 ellipsometer, the recommended range of the angle of incidence φ is 45°÷70°. At angles of incidence φ > 70°, the efficiency of this variant drops sharply. There are two reasons. First, at angles of incidence φ > 70°, the scattering increases noticeably upon reflection of the beam, this leads to a significant increase in the error in determining n_{eff}. Secondly, at large values of the angle of incidence φ, the change in the angle between the beam and the optical axis (if the plane of incidence is not perpendicular to the optical axis!) during the measurement of the dependence $n_{eff} = f(\varphi)$ is very insignificant. And these changes are the smaller, the closer the value of the angle δ to the desired value of the angle *V*. Therefore, the obtained values of n_{eff} will also differ insignificantly. And if we take into account the increased measurement error, then the spread in the values of n_{eff} will be completely random. It is clear that this sharply reduces the sensitivity of such a technique in the range of incidence angles $\varphi > 70°$.

3. Conclusions

Thus, in the third part of the article, which is devoted to the description of the combined ellipsometric method for the complete optical characterization of crystals, two variants of the technique for experimentally determining the orientation of optical axes using ellipsometry are presented. The first variant is based on the search for the circular cross section of the optical indicatrix. As is known, this cross section is perpendicular to the optical axis of the crystal. This is done by successive measurements of the dependence of the effective refractive index on the angle of rotation of the crystal around the normal to the plane under study, $n_{eff} = f(\alpha)$. The implementation of this variant requires the use of modern technological equipment for successive rotations of the investigated plane by 1-1.5° around the desired direction. The second variant is based on the fact that the effective refractive index, n_{eff}, does not depend on the angle of incidence φ of the beam on the plane under study, if the angle between the incident beam and the optical axis does not change. Thus, if measurements are performed on the plane of the optical axes of the crystal, then the problem is reduced to finding a plane that is perpendicular to the optical axis. This is done by successive measurements of the dependence of the effective refractive index on the angle of incidence of the beam on the plane of the optical axes, $n_{eff} = f(\varphi)$. The second variant of this technique was tested by ellipsometric measurements of a $CdWO_4$ crystal made on the plane of the optical axes. It has been established that the angle between the optical axes (the bisector of the angle is the principal axis $\boldsymbol{N_g}$ of the optical indicatrix) is equal to $2V = 92° \pm 1°$. Since $2V > 90°$, the $CdWO_4$ crystal is an optically negative crystal (the optical sign is minus). Thus, the conclusion made in the second part of the article was confirmed that the $CdWO_4$ crystal is a pronounced optically biaxial crystal with an angle between the optical axes close to 90°.

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КОМБІНОВАНА ЕЛІПСОМЕТРИЧНА МЕТОДИКА ПОВНОЇ ОПТИЧНОЇ ХАРАКТЕРИЗАЦІЇ КРИСТАЛІВ. III. ЕКСПЕРИМЕНТАЛЬНЕ ВИЗНАЧЕННЯ ОРІЄНТАЦІЇ ОПТИЧНИХ ОСЕЙ В КРИСТАЛІ

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У третій частині статті, присвяченій опису комбінованої еліпсометричної методики повної оптичної характеризації кристалів, представлені два варіанти методики експериментального визначення орієнтації оптичних осей в кристалі за допомогою еліпсометрії. В основі першого варіанту – пошук колового перерізу оптичної індикатриси, який, як відомо, перпендикулярний оптичній осі кристала. Отже, знайшовши цей переріз, автоматично визначаємо орієнтацію оптичної осі. Виконують цей пошук шляхом послідовних вимірювань залежності ефективного показника заломлення від кута повороту кристала навколо нормалі до досліджуваної площини, $n_{eff} = f(\alpha)$. Практична реалізація цього варіанту потребує використання сучасного технологічного обладнання для послідовних поворотів досліджуваної площини на 1-1.5° навколо середньої головної осі $\boldsymbol{N_m}$ оптичної індикатриси. Другий варіант ґрунтується на тому факті, що ефективний показник заломлення, n_{eff}, не залежить від кута падіння φ променя на досліджувану площину, якщо кут між падаючим променем і оптичною віссю не змінюється. Таким чином, якщо виконувати вимірювання в площині оптичних осей кристала, то завдання зводиться до пошуку площини, яка перпендикулярна оптичній осі. Виконують це шляхом послідовних вимірювань залежності ефективного показника заломлення від кута падіння променя на площину оптичних осей, $n_{eff} = f(\varphi)$. Другий варіант методики був протестований еліпсометричними вимірюваннями кристала $CdWO_4$, виконаними на площині оптичних осей. Встановлено, що кут між оптичними осями, бісектрисою якого є велика головна вісь $\boldsymbol{N_g}$ оптичної індикатриси, дорівнює $2V = 92° \pm 1°$. Оскільки $2V > 90°$, то кристал $CdWO_4$ є оптично негативним кристалом (оптичний знак – мінус). Таким чином, був підтверджений висновок, зроблений у другій частині цієї статті, що кристал $CdWO_4$ – це яскраво виражений оптично двовісний кристал з кутом між оптичними осями близьким до 90°.

Ключові слова: еліпсометрія, оптична індикатриса, головні показники заломлення, одновісні і двовісні кристали.

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