

## DESCRIPTION

TITLE OF THE INVENTION: PRESSURE-SENSITIVE ADHESIVE LAYER,  
METHOD FOR PRODUCING SAME, PRESSURE-SENSITIVE ADHESIVE  
SHEET, PRESSURE-SENSITIVE ADHESIVE LAYER-ATTACHED OPTICAL  
FILM, AND IMAGE DISPLAY DEVICE

## TECHNICAL FIELD

[0001]

The present invention relates to a pressure-sensitive adhesive layer and a method for producing the same. The present invention also relates to a pressure-sensitive adhesive sheet having the pressure-sensitive adhesive layer and a pressure-sensitive adhesive layer-attached optical film. Furthermore, the present invention relates to an image display device using these.

## BACKGROUND ART

[0002]

In a display device such as a liquid crystal display or an organic EL display, a pressure-sensitive adhesive composition is used to bond an optical film such as a polarizing film, a retardation film, a transparent cover member including a cover glass, and other various optical films to another optical film. Thus, by disposing the pressure-sensitive adhesive layer between two optical

films, an optical film laminate having the two optical films is formed. In a display device, the optical film laminate configured as above is disposed such that the optical film is located on a viewing side, for example. This configuration involves a problem that, when outside light enters from the optical film on the viewing side, the entered light is reflected at an interface between the pressure-sensitive adhesive layer and the optical film on a non-viewing side, and returned to the viewing side. This problem becomes prominent particularly when an entry angle of outside light is relatively large.

[0003]

On the other hand, for a backlight unit of an image display device, for example, it has been proposed to use a light-diffusing pressure-sensitive adhesive composition containing a (meth)acrylic-based polymer as a base polymer and light-diffusing fine particles (Patent Document 1).

#### PRIOR ART DOCUMENTS

#### PATENT DOCUMENTS

[0004]

Patent Document 1: JP-A-2014-224964

#### SUMMARY OF THE INVENTION

#### PROBLEMS TO BE SOLVED BY THE INVENTION

[0005]

It is considered that the problem as described above occurs due to a difference in refractive index between the pressure-sensitive adhesive layer and an adherend. For example, between the pressure-sensitive adhesive layer and an optical member (such as an anti-reflection film, a light diffusion film, a light guide film, a prism film, a lens film, a Fresnel lens, a lenticular lens, or a microlens film) using a low refractive index material such as a fluororesin, polysiloxane, low refractive inorganic particles, a porous material, or a hollow material, there occurs a problem in visibility due to an influence of internal reflection of incident light at an interface. Since the problem is considered to be caused by the fact that the refractive index of the optical film is lower than that of the pressure-sensitive adhesive layer, it is considered that the problem can be solved by using a pressure-sensitive adhesive layer having a low refractive index. For example, as a method of forming a pressure-sensitive adhesive layer having a low refractive index with an acrylic pressure-sensitive adhesive, it is conceivable to use fluoroalkyl acrylate (refractive index is around 1.38) as a monomer unit for a general acrylic-based polymer (refractive index is usually 1.47 to 1.52) as a base polymer. However, a pressure-sensitive adhesive layer

having a low refractive index using a base polymer containing the fluoroalkyl acrylate as a monomer unit, has a high surface tension, and it is difficult to secure adhesiveness. In this way, it has been extremely difficult to form a pressure-sensitive adhesive layer of a low refractive index (for example, a refractive index of 1.40 or less) while maintaining the adhesiveness of the pressure-sensitive adhesive layer.

[0006]

On the other hand, although the pressure-sensitive adhesive layer formed from the light-diffusing pressure-sensitive adhesive composition of Patent Document 1 has a light-diffusing function, the light-diffusing fine particles are dispersed in the entire range of the pressure-sensitive adhesive layer, and therefore, it is difficult to secure sufficient adhesiveness with the optical film and the like.

[0007]

It is an object of the present invention to provide a pressure-sensitive adhesive layer, which can effectively suppress internal reflection even when applied to an optical member having a low refractive index such as an anti-reflection film, a light diffusion film, a lens film, a Fresnel lens, a lenticular lens, or a microlens film, and has good adhesiveness and a method for producing the same.

[0008]

Further, it is an object of the present invention to provide a pressure-sensitive adhesive sheet having the pressure-sensitive adhesive layer. Further, it is an object of the present invention to provide a pressure-sensitive adhesive layer-attached optical film having the pressure-sensitive adhesive layer. Furthermore, it is an object of the present invention to provide an image display device having the pressure-sensitive adhesive layer or the pressure-sensitive adhesive layer-attached optical film.

#### MEANS FOR SOLVING THE PROBLEMS

[0009]

As a result of active investigations for solving the problems, the inventors have found the pressure-sensitive adhesive layer and the like described below, so that the invention has been completed.

[0010]

The present invention relates to a pressure-sensitive adhesive layer including a first surface and a second surface on a side opposite to the first surface,

wherein the pressure-sensitive adhesive layer forms a base of a whole of the pressure-sensitive adhesive layer with a pressure-sensitive adhesive composition containing a base polymer, and

the first surface has a first refractive index based on the pressure-sensitive adhesive composition, while a second refractive index of the second surface is lower than the first refractive index of the first surface.

[0011]

In the pressure-sensitive adhesive layer, a difference between the first refractive index of the first surface and the second refractive index of the second surface is preferably 0.02 to 0.45.

[0012]

In the pressure-sensitive adhesive layer, the second refractive index of the second surface is preferably 1.45 or less.

[0013]

In the pressure-sensitive adhesive layer, a low refractive index material having a lower refractive index than the refractive index of the base polymer dispersed on a side of the second surface can be specifically adopted.

[0014]

In the pressure-sensitive adhesive layer, a thickness of a region where the low refractive index material is dispersed is preferably 600 nm or less in a thickness direction from the side of the second surface in the pressure-sensitive adhesive layer.

[0015]

In the pressure-sensitive adhesive layer, the base polymer has a refractive index of 1.40 to 1.55, and the low refractive material preferably has a refractive index of 1.10 to 1.45. And a difference between the refractive index of the base polymer and the refractive index of the low refractive index material is preferably 0.07 to 0.45.

[0016]

The low refractive index material can be particles having an average particle diameter of 10 nm to 150 nm.

[0017]

The low refractive index material can be at least one particle selected from a group consisting of at least one inorganic particle selected from a group consisting of  $\text{MgF}_2$ ,  $\text{CaF}_2$  and  $\text{Na}_3\text{AlF}_6$ , a porous silica particle, a hollow nanosilica particle, and a hollow polymer particle.

[0018]

In the pressure-sensitive adhesive layer, a total light transmittance is preferably 85% or more.

[0019]

In the pressure-sensitive adhesive layer, the second surface preferably has a reflectance of 0.5 to 3.5%.

[0020]

In the pressure-sensitive adhesive layer, a difference in reflectance between the first surface and the second surface is preferably 0.1 to 3.5%.

[0021]

In the pressure-sensitive adhesive layer, a gel fraction is preferably 30 to 95% by weight.

[0022]

In the pressure-sensitive adhesive layer, a storage elastic modulus  $G'$  at 25°C is preferably 0.05 to 0.50 MPa.

[0023]

In the pressure-sensitive adhesive layer, a  $\tan \delta$  peak value is preferably -5 to -50°C when a dynamic viscoelasticity is measured at 1 Hz.

[0024]

The present invention also relates to a method for producing the pressure-sensitive adhesive layer, including:

a step (1) of forming a base pressure-sensitive adhesive layer on a support with a pressure-sensitive adhesive composition containing a base polymer;

a step (2) of providing a dispersion in which a low refractive index material having a lower refractive index than the refractive index of the base polymer is dispersed;

a step (3) of applying the dispersion to a second surface on a side opposite to a first surface on a side of the support in the base pressure-sensitive adhesive layer and penetrating the low refractive index material contained in the dispersion in a thickness direction from the second surface in the base pressure-sensitive adhesive layer; and

a step (4) of drying the pressure-sensitive adhesive layer into which the low refractive index material has penetrated.

[0025]

The present invention also relates to a pressure-sensitive adhesive sheet including: the pressure-sensitive adhesive layer; and a support on one surface or both surfaces of the pressure-sensitive adhesive layer.

[0026]

The present invention also relates to a pressure-sensitive adhesive layer-attached optical film including:

an optical film; and a pressure-sensitive adhesive layer provided on one surface or both surfaces of the optical film,

wherein the pressure-sensitive adhesive layer on the one surface or the both surfaces is the pressure-sensitive adhesive layer, and the optical film is attached to the first surface side of the pressure-sensitive adhesive layer.

[0027]

In the pressure-sensitive adhesive layer-attached optical film, the optical film can be preferably a polarizing film.

[0028]

The present invention also relates to an optical

laminate including: the pressure-sensitive adhesive layer-attached optical film; and a low-refractive-index optical member bonded to the pressure-sensitive adhesive layer of the pressure-sensitive adhesive layer-attached optical film.

[0029]

The present invention also relates to an image display device including the pressure-sensitive adhesive layer, the pressure-sensitive adhesive layer-attached optical film, or the optical laminate.

#### EFFECT OF THE INVENTION

[0030]

The pressure-sensitive adhesive layer of the present invention is different from a diffusion pressure-sensitive adhesive layer in which fine particles are uniformly dispersed in the pressure-sensitive adhesive layer, and has different refractive indexes on both surfaces of one pressure-sensitive adhesive layer having a first surface and a second surface. The pressure-sensitive adhesive layer has, on the first surface side, a first refractive index based on a pressure-sensitive adhesive composition that forms a base of a whole of the pressure-sensitive adhesive layer and has, on the second surface side as the other side, a second refractive index lower than the first

refractive index of the first surface. Thus, since the pressure-sensitive adhesive layer of the present invention has a pressure-sensitive adhesive surface controlled to have a lower refractive index than the refractive index based on the pressure-sensitive adhesive layer, a difference in refractive index between the pressure-sensitive adhesive layer and an optical member (such as an anti-reflection film, a light diffusion film, a light guide film, a prism film, a lens film, a Fresnel lens, a lenticular lens, or a microlens film) formed of a low refractive index material can be adjusted, whereby it is possible to suppress reflection at the interface between the pressure-sensitive adhesive layer and the optical member, and, at the same time, to contribute to improvement of light extraction efficiency. When the second surface of the pressure-sensitive adhesive layer of the present invention is applied to a surface-irregularities portion of a microlens or the like, the surface irregularities can be protected by filling the surface-irregularities portion with the pressure-sensitive adhesive layer. As compared with a case where the surface-irregularities portion includes a void layer, a void can be filled without impairing the light extraction efficiency, and scratches and/or damage to the shape due to vibration and the like during handling or transportation can be suppressed.

Although the second surface of the pressure-sensitive adhesive layer of the present invention has a refractive index adjustment region adjusted to a low refractive index, a low refractive index region can be formed such that a total light transmittance is high without increasing a haze value. Since an adhesive force inherent in the pressure-sensitive adhesive layer is maintained on the first surface of the pressure-sensitive adhesive layer of the present invention, the adhesiveness with a general optical film (such as a polarizing film) is also good. On the other hand, since the pressure-sensitive adhesive composition forms the base on the second surface of the pressure-sensitive adhesive layer controlled to have a low refractive index, the adhesiveness with the optical film formed of a low refractive index material can be secured.

#### BRIEF DESCRIPTION OF THE DRAWINGS

[0031]

Fig. 1 is a cross-sectional view showing an embodiment of a pressure-sensitive adhesive layer of the present invention.

Fig. 2 is a plan view showing a state of a second surface of the pressure-sensitive adhesive layer of the present invention.

Fig. 3 is a schematic view showing steps for

producing the pressure-sensitive adhesive layer of the present invention.

Fig. 4 is a cross-sectional view showing an embodiment of a pressure-sensitive adhesive sheet of the present invention.

Fig. 5 is a cross-sectional view showing an embodiment of a pressure-sensitive adhesive layer-attached optical film of the present invention.

Fig. 6 is a cross-sectional view showing an embodiment of an optical laminate according to the present invention.

#### MODE FOR CARRYING OUT THE INVENTION

[0032]

Hereinafter, a pressure-sensitive adhesive layer and the like of the present invention will be described with reference to the drawings.

[0033]

<Pressure-sensitive adhesive layer>

As shown in Fig. 1, a pressure-sensitive adhesive layer 1 of the present invention has a first surface f1 and a second surface f2 opposite to the first surface f1. In the pressure-sensitive adhesive layer 1, a base (matrix) 1a of the whole of the pressure-sensitive adhesive layer 1 is formed with a pressure-sensitive adhesive composition

containing a base polymer. The first surface f1 has a first refractive index n1, and a second refractive index n2 of the second surface f2 is designed to be lower than the first refractive index n1. Fig. 1 exemplifies a case where in the base 1a, a low refractive index material 2 having a lower refractive index than the refractive index of the base polymer is dispersed (unevenly) on the second surface f2 side.

[0034]

The first refractive index n1 of the first surface f1 corresponds to the refractive index of the pressure-sensitive adhesive layer obtained from the pressure-sensitive adhesive composition forming the base 1a in the pressure-sensitive adhesive layer 1 of the present invention. Therefore, the first refractive index n1 is determined by the pressure-sensitive adhesive composition forming the base 1a. Since the refractive index of the base polymer is substantially the same as the refractive index of the pressure-sensitive adhesive layer 1 of the base 1a of a whole of the pressure-sensitive adhesive layer 1, the first refractive index n1 of the first surface f1 is in general determined by the refractive index of the base polymer. In the pressure-sensitive adhesive composition forming the pressure-sensitive adhesive layer, although will be described later, for example, the refractive index

of the pressure-sensitive adhesive layer formed of a typical acrylic pressure-sensitive adhesive is generally about 1.47 to 1.52. The refractive index of the pressure-sensitive adhesive layer formed of a silicone-based adhesive is generally about 1.40.

[0035]

On the other hand, the second refractive index  $n_2$  of the second surface  $f_2$  is not particularly limited as long as  $n_1 > n_2$  is satisfied in a relationship with the first refractive index  $n_1$  on the first surface  $f_1$ , and the second refractive index  $n_2$  of the second surface  $f_2$  is appropriately determined in consideration of the refractive index of a low refractive index optical film, which serves as an adherend, and the like. However, if a difference ( $n_1 - n_2$ ) between the first refractive index  $n_1$  and the second refractive index  $n_2$  is too large, internal reflection in the pressure-sensitive adhesive layer 1 may occur, so that the difference ( $n_1 - n_2$ ) is preferably adjusted to be 0.02 to 0.45. The difference ( $n_1 - n_2$ ) is more preferably 0.03 to 0.35, and still more preferably 0.03 to 0.25.

[0036]

The second refractive index  $n_2$  of the second surface  $f_2$  is, for example, preferably 1.45 or less from the viewpoint of effectively suppressing internal reflection, and is more preferably 1.4 or less, still more preferably

1.35 or less, and even more preferably 1.3 or less. If the second refractive index  $n_2$  is 1.4 or less, the second surface side of the pressure-sensitive adhesive layer 1 of the present invention may be used as a substitute for an air space. On the other hand, the second refractive index  $n_2$  is preferably 1.25 or more, and more preferably 1.28 or more from the viewpoint of maintaining an adhesive strength. The range of the second refractive index  $n_2$  is lower than the lower limit of the refractive index (generally about 1.47 to 1.52) of the pressure-sensitive adhesive layer formed of a typical acrylic pressure-sensitive adhesive.

[0037]

In the pressure-sensitive adhesive layer 1, as shown in Fig. 1, when the low refractive index material 2 is dispersed on the second surface f2 side, the refractive index of the base polymer in the pressure-sensitive adhesive composition forming the base 1a is preferably 1.40 to 1.55, and the refractive index of the low refractive index material 2 dispersed on the second surface a2 side in the base 1a is preferably 1.10 to 1.45. A difference between the refractive index of the base polymer and the refractive index of the low refractive index material 2 is preferably 0.07 to 0.45. The refractive index of the base polymer is more preferably 1.40 to 1.52 and still more

preferably 1.40 to 1.50. The refractive index of the low refractive index material 2 is more preferably 1.14 to 1.42 and still more preferably 1.18 to 1.40. The difference between the refractive index of the base polymer and the refractive index of the low refractive index material 2 is more preferably 0.07 to 0.35 and still more preferably 0.10 to 0.30. The low refractive index material 2 can lower the refractive index with a lower addition amount when the refractive index is lower. On the other hand, the difference between the refractive index of the base polymer (pressure-sensitive adhesive layer 1a) and the refractive index of the low refractive index material 2 increases, and scattering (haze) tends to occur; therefore, it is preferable to adjust the refractive index difference so that the refractive index difference does not become too large. The refractive index can be shown as a refractive index value of a D-line measured by a spectroscopic ellipsometry method in an environment of 23°C when the material is a single layer film.

[0038]

As the low refractive index material 2, particles having an average particle diameter of 10 nm to 150 nm can be used. Even when the particles having an average particle diameter in the above range are dispersed on the second surface f2 side of the pressure-sensitive adhesive

layer 1, the particles are preferable for maintaining a low haze of the pressure-sensitive adhesive layer 1 and a high total light transmittance. The average particle diameter is preferably 20 nm to 100 nm and more preferably 20 nm to 90 nm. The average particle diameter of the particles is a value measured by a particle size distribution analyzer by a dynamic light scattering method.

[0039]

Examples of the low refractive index material 2 include  $\text{MgF}_2$  (refractive index 1.38),  $\text{CaF}_2$  (refractive index 1.43: fluorite), and  $\text{Na}_3\text{AlF}_6$  (refractive index 1.34: sodium hexafluoroaluminate (cryolite)). These materials (for example, particles) can be used alone or in combination of two or more.

[0040]

As the low refractive index material 2, for example, hollow particles can be used. The hollow particles may be either inorganic particles or polymer particles. Since the hollow particles have a void space with a low refractive index in the particles, the refractive index of the hollow particles is lower than the refractive index of a component forming the hollow particles. For example, although silica has a refractive index of 1.46, hollow nanosilica particles (refractive index 1.24, trade name: Throughrear 5320, manufactured by JGC Catalysts and Chemicals Ltd., particle

diameter 75 nm) and porous silica particles can be used as a low refractive index material. In addition, hollow polymer fine particles (refractive index 1.32, trade name: Techpolymer NH, manufactured by Sekisui Kasei Co., Ltd., product number XX-255AA, particle diameter 80 nm, hollow ratio 39%) can be exemplified. When the hollow particles are provided in a low refractive surface treatment layer, there are problems of strength and scratch resistance because the hollow particles are hollow materials. However, since the hollow particles (low refractive index material 2) in the present invention are added (impregnated) to the pressure-sensitive adhesive layer 1, the hollow particles can be applied without considering the problems of strength and scratch resistance.

[0041]

As the low refractive index material 2, a fluoroalkyl group-containing oligomer, an oligomer of polysiloxane resin, or the like can be used.

[0042]

The thickness of the pressure-sensitive adhesive layer 1 is not particularly limited, and is usually 5  $\mu\text{m}$  to 500  $\mu\text{m}$ , preferably 10  $\mu\text{m}$  to 400  $\mu\text{m}$ , and more preferably 10  $\mu\text{m}$  to 350  $\mu\text{m}$ . Further, in Fig. 1, a region where the low refractive index material 2 is dispersed in the pressure-sensitive adhesive layer 1 is represented by a thickness T

from the second surface f2 side. The thickness T is appropriately designed according to the thickness of the pressure-sensitive adhesive layer, and is usually preferably 600 nm or less, more preferably 300 nm or less, and still more preferably 200 nm or less. In order to effectively suppress internal reflection when the thickness T is applied to an optical film having a low refractive index, the thickness T is preferably 10 nm or more, more preferably 15 nm or more, and still more preferably 20 nm or more.

[0043]

In the pressure-sensitive adhesive layer 1, a region where the low refractive index material 2 relating to the thickness T is dispersed has an irregular uneven shape in relation to the base (matrix) 1a. However, in the present invention, the thickness T is determined by averaging a measurement value of a depth of the uneven shape.

[0044]

The low refractive index material 2 is distributed on the second surface f2 side in an individually dispersed state or in a partially aggregated state. As described in connection with Fig. 1, a boundary between the region where the low refractive index material 2 is dispersed and the base 1a in which the low refractive index material 2 is not dispersed has an irregular uneven shape. However, in the

measurement of the thickness  $T$ , a depth range at each measurement position where 90% of the low refractive index material 2 exist is determined as a measurement value of the thickness  $T$  at the measurement position, and the measurement values at the plurality of measurement positions are averaged.

[0045]

Fig. 2 is a plan view showing a state of the second surface  $f2$  of the pressure-sensitive adhesive layer 1. As shown in Fig. 2, the second surface  $f2$  is formed in a sea-island structure where the low refractive index material 2 is dispersed in the base 1a in the form of islands and show a region of the base 1a and a region of the low refractive index material 2. An area ratio of the low refractive index material 2 on the second surface  $f2$  is preferably in a range of 30 to 99%. The area ratio is calculated by measuring an area of the low refractive index material 2 in each of a plurality of square regions having a side length of 10  $\mu\text{m}$  to 200  $\mu\text{m}$  to obtain a ratio of the area of the low refractive index material 2 to the entire area of the square region, and averaging the area ratios measured in the plurality of square regions.

[0046]

A ratio of the low refractive index material 3 in the pressure-sensitive adhesive layer 1 is not particularly

limited as long as the first refractive index  $n_1$  on the first surface  $f_1$  and the second refractive index  $n_2$  of the second surface  $f_2$  satisfy the relationship of  $n_1 > n_2$ .

[0047]

The total light transmittance of the whole of the pressure-sensitive adhesive layer 1 of the present invention is preferably 85% or more, more preferably 88% or more, and still more preferably 90% or more. The higher the total light transmittance of the pressure-sensitive adhesive layer 1, the more preferable. A haze value is preferably 1.5% or less, more preferably 1% or less, and still more preferably 0.8% or less. The lower the haze value of the pressure-sensitive adhesive layer 1, the more preferable. The total light transmittance and haze value of the whole of the pressure-sensitive adhesive layer 1 are values measured according to JIS K7361.

[0048]

A reflectance of the second surface of the pressure-sensitive adhesive layer 1 of the present invention is preferably 0.5 to 3.5%. The reflectance of the second surface of the pressure-sensitive adhesive layer 1 of the present invention is lower than the reflectance of the first surface, and the reflectance of the second surface that can control the internal reflection to be small even in relation to the low refractive index material is

preferably 0.5 to 3.0% and more preferably 0.5 to 2.5%. A difference in reflectance between the first surface and the second surface of the pressure-sensitive adhesive layer 1 of the present invention is preferably 0.1 to 3.5%.

[0049]

In the pressure-sensitive adhesive layer 1 of the present invention, in the above description, it is assumed that the second refractive index  $n_2$  of the second surface  $f_2$  is designed to be lower than the first refractive index  $n_1$ . However, the pressure-sensitive adhesive layer 1 of the present invention can be specified by the relationship of the refractive indexes of both surfaces and, in addition, can be grasped as an invention characterized in that the reflectance of the second surface  $f_2$  is lower than the reflectance of the first surface  $f_1$ .

[0050]

The pressure-sensitive adhesive layer 1 of the present invention preferably has a gel fraction of 30 to 95% by weight. The gel fraction is preferably 30 to 90% by weight, more preferably 35 to 90% by weight, and still more preferably 40 to 90% by weight. When the gel fraction of the pressure-sensitive adhesive layer 1 is within the above range, a stress relaxation property is provided, and this is a preferable embodiment for the purpose of securing followability to unevenness. The gel fraction relates to

the base 1a in the pressure-sensitive adhesive layer 1 and does not include the low refractive index material 2.

[0051]

<Measurement of gel fraction of pressure-sensitive adhesive layer>

About 0.2 g of the pressure-sensitive adhesive layer (before permeation of the low refractive index material) was scraped to prepare a sample 1. The sample 1 was wrapped with a Teflon (registered trademark) film (trade name "NTF1122", manufactured by Nitto Denko Corporation) having a diameter of 0.2  $\mu\text{m}$ , then strapped with a kite string, and used as a sample 2. The weight of the sample 2 before being subjected to the following test was measured, which was taken as a weight A. The weight A is a total weight of the sample 1 (pressure-sensitive adhesive layer), the Teflon (registered trademark) film, and the kite string. A total weight of the Teflon (registered trademark) film and the kite string was defined as a weight B. Then, the sample 2 was placed in a 50 ml container filled with ethyl acetate and left standing at 23°C for 1 week. Thereafter, the sample 2 was taken out from the container, and dried in a dryer at 130°C for 2 hours to remove ethyl acetate, and then the weight of the sample 2 was measured. The weight of the sample 2 after being subjected to the above test was measured, which was taken

as a weight C. Then, the gel fraction (% by weight) was calculated from the following formula.

$$\text{Gel fraction (wt\%)} = (C - B) / (A - B) \times 100$$

[0052]

The pressure-sensitive adhesive layer 1 of the present invention preferably has a storage elastic modulus  $G'$  at 25°C of 0.05 to 0.50 MPa. The storage elastic modulus  $G'$  is preferably 0.06 to 0.45 MPa, more preferably 0.07 to 0.40 MPa, and still more preferably 0.08 to 0.35 MPa. When the storage elastic modulus  $G'$  of the pressure-sensitive adhesive layer 1 is within the above range, this is a preferable embodiment for the purpose of protecting image display devices (LCD and OLED terminals, etc.) that are becoming thinner and securing dimensional stability during cutting.

[0053]

The pressure-sensitive adhesive layer 1 of the present invention preferably has a  $\tan \delta$  peak value (glass transition temperature) of -5 to -50°C when a dynamic viscoelasticity is measured at 1 Hz. The  $\tan \delta$  peak value is preferably -7 to -50°C, more preferably -9 to -45°C, and still more preferably -10 to -40°C. When the  $\tan \delta$  peak value of the pressure-sensitive adhesive layer 1 is within the above range, this is a preferable embodiment for the purpose of securing resistance to drop impact of the image

display device (mobile terminal, etc.).

[0054]

<Measurement of storage elastic modulus  $G'$  and  $\tan \delta$  peak value of pressure-sensitive adhesive layer>

A plurality of pressure-sensitive adhesive layers were stacked to prepare a test sample having a thickness of about 2 mm. The test sample was stamped into a disk having a diameter of 7.9 mm and sandwiched between parallel plates, and the dynamic viscoelasticity was measured under the following conditions using "Advanced Rheometric Expansion System (ARES)" manufactured by Rheometric Scientific, Inc. The storage elastic modulus  $G'$  and the  $\tan \delta$  peak value of the pressure-sensitive adhesive layer at 25°C were read from the measurement results.

(Measurement condition)

Frequency: 1 Hz

Deformation mode: twisting

Measurement temperature: -70°C to 150°C

Rate of temperature increase: 5°C/min

[0055]

Next, a method for producing the pressure-sensitive adhesive layer of the present invention will be described with reference to Fig. 3.

[0056]

First, in a step (1), a base pressure-sensitive

adhesive layer 1' is formed on a support S with a pressure-sensitive adhesive composition containing a base polymer. The base pressure-sensitive adhesive layer 1' forms the base 1a in the pressure-sensitive adhesive layer 1 to be obtained. In the base pressure-sensitive adhesive layer 1', the support 1 side is the first surface f1', and the opposite side is the second surface f2'. A method of forming the base pressure-sensitive adhesive layer 1' is not particularly limited, and the base pressure-sensitive adhesive layer 1' can be formed by a method usually used in this field. Specifically, the base pressure-sensitive adhesive layer 1' can be formed by applying the pressure-sensitive adhesive composition to one surface of the support S and drying a coating film formed from the pressure-sensitive adhesive composition, or can be formed by irradiation with active energy rays such as ultraviolet rays.

[0057]

The support S is not particularly limited, and, for example, various base materials such as a release film and a transparent resin film base material can be used.

[0058]

Examples of the material for forming the release film include a resin film such as a polyethylene, polypropylene, polyethylene terephthalate, and polyester film; a porous

material such as paper, cloth and nonwoven fabric; and an appropriate thin sheet such as a net, a foamed sheet, a metal foil, and a laminate thereof. In particular, a resin film is preferably used, because of its good surface smoothness. If necessary, the release film may be subjected to release and antifouling treatment or to antistatic treatment.

[0059]

On the other hand, in a step (2), a dispersion 10 in which the low refractive index material 2 having a lower refractive index than that of the base polymer used in the pressure-sensitive adhesive composition is dispersed is provided (not shown). As a dispersion medium used in the dispersion, a material that can disperse the low refractive index material 2 and that can penetrate into the base pressure-sensitive adhesive layer 1' is used. The dispersion medium is selected in time according to the type of the low refractive index material and the type of the pressure-sensitive adhesive composition forming the base pressure-sensitive adhesive layer. The concentration of the low refractive index material in the dispersion medium is preferably adjusted to, for example, 0.1 to 10% by weight.

[0060]

Examples of the dispersion medium include alcohols

such as methanol, ethanol, isopropyl alcohol, 1-propanol, n-butanol, 2-butanol, cyclohexanol, t-butyl alcohol, glycerin, ethylene glycol, 2-methyl-2,4-pentadiol, phenol, and parachlorophenol; ketones such as acetone, methyl ethyl ketone, methyl isobutyl ketone, cyclohexanone, cyclopentanone, 2-pentanone, 2-hexanone, and 2-heptanone; ethers such as diethyl ether, tetrahydrofuran, dioxane, and anisole; esters such as ethyl acetate, butyl acetate, and methyl lactate; aromatic hydrocarbons such as benzene, toluene, and xylene; aliphatic hydrocarbons such as n-hexane and cyclohexane; amides such as dimethylformamide and dimethylacetamide; and cellosolves such as methyl cellosolve, ethyl cellosolve, and methyl cellosolve acetate. These dispersion media can be used alone or in combination of two or more. The above solvents are merely examples, and the solvents used in the present invention are not limited to these.

[0061]

Then, as a step (3), the second surface  $f2'$  in the base pressure-sensitive adhesive layer  $1'$  is coated with the dispersion 10, and the low refractive index material 2 contained in the dispersion 10 is penetrated in a thickness direction from the second surface  $f2'$  in the base pressure-sensitive adhesive layer  $1'$ . (3)-1 in Fig. 3 shows a state immediately after the dispersion 10 is applied to the base

pressure-sensitive adhesive layer 1', and (3)-2 shows a state where the low refractive index material 2 has penetrated into the base pressure-sensitive adhesive layer 1'. The side of the second surface f2' of the base pressure-sensitive adhesive layer 1' is swollen by the dispersion medium of the dispersion 10, and in the process, the low refractive index material 2 in the dispersion 10 penetrates into the base pressure-sensitive adhesive layer 1'.

[0062]

Then, in a step (4), the base pressure-sensitive adhesive layer 1' into which the low refractive index material 2 has penetrated is dried. The pressure-sensitive adhesive layer 1 shown in Fig. 1 can be obtained by evaporating the dispersion medium of the dispersion 10 that has penetrated into the base pressure-sensitive adhesive layer 1' in a drying step. This state is shown in (4) in Fig. 3. The conditions of the drying step are determined according to the type of dispersion medium.

[0063]

The region (thickness T) where the low refractive index material 2 is dispersed in the pressure-sensitive adhesive layer 1 is determined by a relationship between the pressure-sensitive adhesive composition forming the base pressure-sensitive adhesive layer 1' and the

dispersion medium of the dispersion 10. The dispersion medium may be appropriately selected to enable the infiltration depth to become the aforementioned value. The amount of the dispersion to be applied is appropriately set so as to obtain a desired thickness T.

[0064]

As the method of applying the dispersion, for example, an appropriate method such as roll coating, kiss roll coating, gravure coating, reverse coating, roll brush coating, spray coating, dip roll coating, bar coating, knife coating, air knife coating, curtain coating, lip coating, or coating with a die coater can be used. The thickness T can be controlled by the method of applying the dispersion, the concentration of the dispersion, the application amount, and the like.

[0065]

<Pressure-sensitive adhesive composition>

The pressure-sensitive adhesive composition containing the base polymer forming the base (matrix) 1a of the pressure-sensitive adhesive layer 1 of the present invention will be described.

[0066]

For the pressure-sensitive adhesive composition, a transparent adherent material usable in optical applications is preferably used. As the pressure-sensitive

adhesive composition, for example, it is possible to use one or more appropriately selected from the group consisting of an acrylic-based pressure-sensitive adhesive, a rubber-based pressure-sensitive adhesive, a silicone-based pressure-sensitive adhesive, a polyester-based pressure-sensitive adhesive, a urethane-based pressure-sensitive adhesive, an epoxy-based pressure-sensitive adhesive, and a polyether-based adhesive. From the viewpoint of transparency, processability, durability, etc., it is preferable to use an acrylic pressure-sensitive adhesive. The base-polymer may be used a base-polymer which is adapted for a kind of a pressure-sensitive-adhesive composition. In the present invention, the pressure-sensitive-pressure-sensitive adhesive including, as its base polymer, a (meth)acrylic-based polymer.

[0067]

The acrylic-based pressure-sensitive-pressure-sensitive adhesive preferably includes, for example, a partially polymerized product of one or more monomer components which include an alkyl (meth)acrylate, and/or a (meth)acrylic-based polymer yielded from the monomer component(s). The above-mentioned acrylic-based pressure-sensitive-pressure-sensitive adhesive includes a partially polymerized product of one or more monomer component(s) which include an alkyl (meth)acrylate, and/or a

(meth)acrylic-based polymer yielded from the monomer component(s).

[0068]

The alkyl (meth)acrylate may be the alkyl (meth)acrylate having a linear or branched alkyl group of 1 to 24 carbon atoms, among them, preferred is an alkyl (meth)acrylate having an alkyl group of 1 to 9 carbon atoms, more preferred is an alkyl (meth)acrylate having a branched alkyl group of 4 to 9 carbon atoms. These alkyl (meth)acrylates are each favorable since the resultant pressure-sensitive-pressure-sensitive adhesive layer easily takes a balance between its adhesive properties. Specific examples of the alkyl (meth)acrylate having a branched alkyl group of 4 to 9 carbon atoms, include n-butyl (meth)acrylate, s-butyl (meth)acrylate, t-butyl (meth)acrylate, isobutyl (meth)acrylate, n-pentyl (meth)acrylate, isopentyl (meth)acrylate, isohexyl (meth)acrylate, isoheptyl (meth)acrylate, 2-ethylhexyl (meth)acrylate, isooctyl (meth)acrylate, and isononyl (meth)acrylate. These may be used singly or in any combination of two or more.

[0069]

In the present invention, the proportion of the above-mentioned alkyl (meth)acrylate having an alkyl group of 1 to 24 carbon atoms at an ester end, is preferably 40%

or more by weight, more preferably 50% or more by weight, even more preferably 60% or more by weight of the whole of one or more monofunctional monomer components from which the (meth)acrylic-based polymer is made.

[0070]

The above-mentioned monomer component(s) may include, as one of the monofunctional monomer components, a copolymerizable monomer other than the alkyl (meth)acrylate. The copolymerizable monomer is usable as the rest of the monomer component(s) that is other than the alkyl (meth)acrylate.

[0071]

As the copolymerizable monomer, the monomer component(s) may include, for example, a cyclic nitrogen-containing monomer. It is allowable to use, as the cyclic nitrogen-containing monomer, a monomer having a polymerizable functional group having an unsaturated double bond, such as a (meth)acryloyl group or vinyl group, and further having a cyclic nitrogen structure without any especial limitation. The cyclic nitrogen structure is preferably a cyclic structure having therein one or more nitrogen atoms. Examples of the cyclic nitrogen-containing monomer include lactam-based vinyl monomers such as N-vinylpyrrolidone, N-vinyl- $\epsilon$ -caprolactam, and methylvinylpyrrolidone; vinyl monomers each having a

nitrogen-containing heterocyclic ring such as vinylpyridine, vinylpiperidone, vinylpyrimidine, vinylpiperazine, vinylpyrazine, vinylpyrrole, vinylimidazole, vinyloxazole, and vinylmorpholine. Other examples thereof include (meth)acrylic monomers each containing a heterocyclic ring such as a morpholine ring, a piperidine ring, a pyrrolidine ring, or a piperazine ring. Specific examples thereof include N-acryloylmorpholine, N-acryloylpiperidine, N-methacryloylpiperidine, and N-acryloylpyrrolidine. Among these cyclic nitrogen-containing monomers, lactam-based vinyl monomers are preferred.

[0072]

In the present invention, the proportion of the cyclic nitrogen-containing monomer is preferably from 0.5 to 50% by weight, more preferably from 0.5 to 40% by weight, even more preferably from 0.5 to 30% by weight of the whole of the monofunctional monomer component(s) from which the (meth)acrylic-based polymer is made.

[0073]

The monomer component(s) used in the present invention may include, as one of the monofunctional monomer components, a hydroxyl-group-containing monomer. It is allowable to use, as the hydroxyl-group-containing monomer, a monomer having a polymerizable functional group having an

unsaturated double bond, such as a (meth)acryloyl group or vinyl group, and further having a hydroxyl group with especial limitation. Examples of the hydroxyl-group-containing monomer include hydroxyalkyl (meth)acrylates such as 2-hydroxyethyl (meth)acrylate, 2-hydroxypropyl (meth)acrylate, 2-hydroxybutyl (meth)acrylate, 3-hydroxypropyl (meth)acrylate, 4-hydroxybutyl (meth)acrylate, 6-hydroxyhexyl (meth)acrylate, 8-hydroxyoctyl (meth)acrylate, 10-hydroxydecyl (meth)acrylate, and 12-hydroxylauryl (meth)acrylate; and hydroxyalkylcycloalkane (meth)acrylates such as (4-hydroxymethylcyclohexyl)methyl (meth)acrylate. Other examples thereof include hydroxyethyl (meth)acrylamide, allyl alcohol, 2-hydroxyethyl vinyl ether, 4-hydroxybutyl vinyl ether, and diethylene glycol monovinyl ether. These monomers may be used singly or in combination. Among them, hydroxyalkyl (meth)acrylates are preferred.

[0074]

In the present invention, since the pressure-sensitive-pressure-sensitive adhesive composition is heightened in adhering strength and cohesive strength, the proportion of the hydroxyl-group-containing monomer is preferably 1% or more by weight, more preferably 2% or more by weight, even more preferably 3% or more by weight of the whole of the monofunctional monomer component(s), from

which the (meth)acrylic-based polymer is made. However, if the proportion of the hydroxyl-group-containing monomer is too large, the pressure-sensitive-pressure-sensitive adhesive layer may be hardened to be lowered in adhering strength. Moreover, the pressure-sensitive adhesive may become too high in viscosity or may be gelatinized. Thus, the proportion of the hydroxyl-group-containing monomer is preferably 30% or less by weight, more preferably 27% or less by weight, even more preferably 25% or less by weight of the whole of the monofunctional monomer component(s), from which the (meth)acrylic-based polymer is made.

[0075]

The monomer component(s), from which the (meth)acrylic-based polymer is made, may contain, as one of the monofunctional monomers, any other functional-group-containing monomer. The other functional-group-containing monomer is, for example, a carboxyl-group-containing monomer or a monomer having a cyclic ether group.

[0076]

It is allowable to use, as the carboxyl-group-containing monomer, a monomer having a polymerizable functional group having an unsaturated double bond, such as a (meth)acryloyl group or vinyl group, and further having a carboxyl group without any especial limitation. Examples of the carboxyl-group-containing monomer include

(meth)acrylic acid, carboxyethyl (meth)acrylate, carboxypentyl (meth)acrylate, itaconic acid, maleic acid, fumaric acid, crotonic acid, and isocrotonic acid. These monomers are usable singly or in combination. Itaconic acid and maleic acid are each usable in the form of an anhydride thereof. Among them, acrylic acid and methacrylic acid are preferred, and acrylic acid is particularly preferred. For the monomer component(s) used in the production of the (meth)acrylic-based polymer in the present invention, the carboxyl-group-containing monomer is arbitrarily usable. However, the carboxyl-group-containing monomer may not be used.

[0077]

It is allowable to use, as the monomer having a cyclic ether group, a monomer having a polymerizable functional group having an unsaturated double bond, such as a (meth)acryloyl group or vinyl group, and further having a cyclic ether group such as an epoxy group or oxetane group without any especial limitation. Examples of the epoxy-group-containing monomer include glycidyl (meth)acrylate, 3,4-epoxycyclohexylmethyl (meth)acrylate, and 4-hydroxybutyl (meth)acrylate glycidyl ether. Examples of the oxetane-group-containing monomer include 3-oxetanylmethyl (meth)acrylate, 3-methyl-oxetanylmethyl (meth)acrylate, 3-ethyl-oxetanylmethyl (meth)acrylate, 3-

butyl-oxetanylmethyl (meth)acrylate, and 3-hexyl-oxetanylmethyl (meth)acrylate. These monomers may be used singly or in combination.

[0078]

In the present invention, the proportion of the carboxyl-group-containing monomer or the monomer having a cyclic ether group is preferably 30% or less by weight, more preferably 27% or less by weight, even more preferably 25% or less by weight of the whole of the monofunctional monomer component(s), from which the (meth)acrylic-based polymer is made.

[0079]

Among the monomer components, from which the (meth)acrylic-based polymer in the present invention is made, the copolymerizable monomer is, for example, an alkyl (meth)acrylate represented by  $\text{CH}_2=\text{C}(\text{R}^1)\text{COOR}^2$  wherein  $\text{R}^1$  represents hydrogen or a methyl group, and  $\text{R}^2$  represents a substituted alkyl group of 1 to 3 carbon atoms, or a cyclic alkyl group.

[0080]

About the substituted alkyl group of 1 to 3 carbon atoms as  $\text{R}^2$ , its substituent is preferably an aryl group of 3 to 8 carbon atoms or an aryloxy group of 3 to 8 carbon atoms. The aryl group is not limited, and is preferably a phenyl group.

[0081]

Examples of the monomer represented by  $\text{CH}_2=\text{C}(\text{R}^1)\text{COOR}^2$  include phenoxyethyl (meth)acrylate, benzyl (meth)acrylate, cyclohexyl (meth)acrylate, 3,3,5-trimethylcyclohexyl (meth)acrylate, and isobornyl (meth)acrylate. These monomers may be used singly or in combination.

[0082]

In the present invention, the proportion of the (meth)acrylate represented by  $\text{CH}_2=\text{C}(\text{R}^1)\text{COOR}^2$  may be 50% or less by weight of the whole of the monofunctional monomer component(s), from which the (meth)acrylic-based polymer is made, and is preferably 45% or less by weight, more preferably 40% or less by weight, even more preferably 35% or less by weight thereof.

[0083]

The following may also be used as other copolymerizable monomer: vinyl acetate, vinyl propionate, styrene,  $\alpha$ -methyl styrene; glycol acrylate-based monomers such as polyethylene glycol (meth)acrylate, polypropylene glycol (meth) acrylate, methoxy ethylene glycol (meth)acrylate, and methoxy polypropylene glycol (meth)acrylate; acrylate-based monomers such as tetrahydrofurfuryl (meth)acrylate, fluorine (meth)acrylate, silicone (meth)acrylate, and 2-methoxyethyl acrylate; and amide-group-containing monomers, amino-group-containing

monomers, imide-group-containing monomers, N-acryloylmorpholine, and vinyl ether monomers. Furthermore, the following may also be used as the copolymerizable monomer: terpene (meth)acrylate, dicyclopentanyl (meth)acrylate, or any other monomer having a cyclic structure.

[0084]

Furthermore, the copolymerizable monomer may be, for example, a silicon-atom-containing silane-based monomer. Examples of the silane-based monomer include 3-acryloxypropyltriethoxysilane, vinyltrimethoxysilane, vinyltriethoxysilane, 4-vinylbutyltrimethoxysilane, 4-vinylbutyltriethoxysilane, 8-vinyl-octyltrimethoxysilane, 8-vinyloctyltriethoxysilane, 10-methacryloyloxydecyltrimethoxysilane, 10-acryloyloxydecyltrimethoxysilane, 10-methacryloyloxydecyltriethoxysilane, and 10-acryloyloxydecyltriethoxysilane.

[0085]

The monomer component(s), from which the (meth)acrylic-based polymer in the present invention is made, may optionally include a polyfunctional monomer beside the above-exemplified monofunctional monomers, so as to adjust the cohesive strength of the pressure-sensitive adhesive.

[0086]

The polyfunctional monomer is a monomer having at least two polymerizable functional groups each having an unsaturated double bond such as a (meth)acryloyl group or vinyl group. Examples thereof include ester compounds of a polyhydric alcohol with (meth)acrylic acid such as (poly)ethylene glycol di(meth)acrylate, (poly)propylene glycol di(meth)acrylate, neopentyl glycol di(meth)acrylate, pentaerythritol di(meth)acrylate, pentaerythritol tri(meth)acrylate, dipentaerythritol hexa(meth)acrylate, 1,2-ethylene glycol di(meth)acrylate, 1,6-hexanediol di(meth)acrylate, 1,12-dodecanediol di(meth)acrylate, trimethylolpropane tri(meth)acrylate, and tetramethylolmethane tri(meth)acrylate; and allyl (meth)acrylate, vinyl (meth)acrylate, divinylbenzene, epoxy acrylate, polyester acrylate, urethane acrylate, butyl di(meth)acrylate, and hexyl di(meth)acrylate. Among them, the following is preferably usable: trimethylolpropane tri(meth)acrylate, hexanediol di(meth)acrylate, or dipentaerythritol hexa(meth)acrylate. Such polyfunctional monomers may be used singly or in combination of two or more thereof.

[0087]

The use amount of the polyfunctional monomer(s), which is varied in accordance with the molecular weight

thereof, the number of functional groups thereof and others, is preferably 3 parts or less by weight, more preferably 2 parts or less by weight, even more preferably 1 part or less for 100 parts by weight of the whole of the monofunctional monomer(s). The lower limit thereof is not particularly limited, and is preferably 0 parts or more by weight, more preferably 0.001 part or more by weight. When the use amount of the polyfunctional monomer(s) is in any one of these ranges, the pressure-sensitive-pressure-sensitive adhesive composition can be improved in adhering strength.

[0088]

For the production of the (meth)acrylic-based polymer, a known producing method is appropriately selectable, examples thereof including solution polymerization, ultraviolet (UV) polymerization, other radial ray polymerizations, bulk polymerization, emulsion polymerization, and various other radical polymerizations. The resultant (meth)acrylic-based polymer may be any one of a random copolymer, a block copolymer, a graft copolymer and others.

[0089]

In the present invention, a partially polymerized product of the monomer component(s) is also preferably usable.

[0090]

For the production of the (meth)acrylic-based polymer, a known producing method is appropriately selectable, examples thereof including solution polymerization, ultraviolet (UV) polymerization, other radial ray polymerizations, bulk polymerization, emulsion polymerization, and various other radical polymerizations. The resultant (meth)acrylic-based polymer may be any one of a random copolymer, a block copolymer, a graft copolymer and others.

[0091]

When the (meth)acrylic-based polymer is produced by a radical polymerization, the polymerization can be conducted by adding, to the monomer component(s), a polymerization initiator, a chain transfer agent, an emulsifier, and others that are used in the radical polymerization. The polymerization initiator, the chain transfer agent, the emulsifier, and the others, which are used in the radical polymerization, are not particularly limited, and may be appropriately selected to be used. The weight-average molecular weight of the (meth)acrylic-based polymer is controllable in accordance with the respective use amounts of the polymerization initiator and the chain transfer agent, and conditions for the reaction. Correspondingly to the species of these substances, the use amounts are

appropriately adjusted.

[0092]

For example, in the solution polymerization, a used polymerization solvent is, for example, ethyl acetate or toluene. In a specific example of the solution polymerization, reaction is conducted in the presence of an added polymerization initiator under a gas flow of an inert gas such as nitrogen usually under reaction conditions of a temperature of about 50 to 70°C and a period of about 5 to 30 hours.

[0093]

When the (meth)acrylic-based polymer is produced through radiation polymerization, the (meth)acryl polymer can be produced by radiation polymerization, in which radiation, such as electron beams or UV rays, is applied to the monomer component. When UV polymerization is performed, a photopolymerization initiator is preferably added to the monomer component, which is advantageous in that the polymerization time can be reduced.

[0094]

(Silane Coupling Agent)

Furthermore, the pressure-sensitive-pressure-sensitive adhesive composition of the present invention may contain a silane coupling agent. The blend amount of the silane coupling agent is preferably 1 part or less by

weight, more preferably from 0.01 to 1 part by weight, even more preferably from 0.02 to 0.6 part by weight for 100 parts by weight of the base-polymer (for example, the above (meth)acrylic-based polymer).

[0095]

(Crosslinking Agent)

The pressure-sensitive-pressure-sensitive adhesive composition of the present invention may contain a crosslinking agent. Examples of the crosslinking agent include isocyanate-based crosslinking agents, epoxy-based crosslinking agents, silicone-based crosslinking agents, oxazoline-based crosslinking agents, aziridine-based crosslinking agents, silane-based crosslinking agents, alkyl-etherified melamine-based crosslinking agents, metal chelate-based crosslinking agents, and peroxide crosslinking agents. These crosslinking agents may be used singly or in any combination of two or more thereof. Among them, isocyanate-based crosslinking agents are preferred.

[0096]

The crosslinking agents may be used singly or in the form of a mixture of two or more thereof. The content of the whole of the crosslinking agent(s) is preferably 5 parts or less by weight, more preferably from 0.01 to 5 parts by weight, even more preferably from 0.01 to 4 parts by weight, in particular preferably from 0.02 to 3 parts by

weight for 100 parts by weight of the monofunctional monomer component(s), from which the (meth)acrylic-based polymer is made.

[0097]

(Other additives)

In addition to the above components, the pressure-sensitive adhesive composition of the present invention may contain appropriate additives depending on the application. Examples of additives include viscosity modifiers, release modifiers, tackifiers (including ones that are solid, semisolid, or liquid at ambient temperature and mainly contain materials such as rosin derivative resins, polyterpene resins, petroleum resins, and oil-soluble phenolic resins), plasticizers, softening agents, pigments, colorants (such as pigments and dyes), pH adjusters (acid or base), rust preventives, age inhibitors, antioxidants, light stabilizers, and ultraviolet absorbers.

[0098]

The pressure-sensitive adhesive sheet of the present invention has the pressure-sensitive adhesive layer 1 and a support on one surface or both surfaces of the pressure-sensitive adhesive layer 1. Fig. 4 shows a case where the pressure-sensitive adhesive layer 1 has a support 3a on the first surface f1 and a support 3b on the second surface f2. As the supports 3a and 3b, the same support S as that used

in the pressure-sensitive adhesive layer 1 shown in Fig. 3 can be used. As the support 3a, the support S used in the method for producing the pressure-sensitive adhesive layer 1 shown in Fig. 3 can be used as it is. The support 3b can be appropriately provided on the second surface f2 of the pressure-sensitive adhesive layer 1 after the pressure-sensitive adhesive layer 1 is produced by the producing method shown in Fig. 3.

[0099]

The pressure-sensitive adhesive layer-attached optical film A of the present invention has the optical film 4 and the pressure-sensitive adhesive layer 1 provided on one surface or both surfaces of the optical film 4. The pressure-sensitive adhesive layer 1 is provided on either one surface or both surfaces of the optical film 4. In the pressure-sensitive adhesive layer 1, the first surface f1 side of the pressure-sensitive adhesive layer 1 is provided on the optical film 4. When the pressure-sensitive adhesive layer 1 is provided on one surface of the optical film 4, a usual pressure-sensitive adhesive layer may be provided on the other surface. In Fig. 5, the pressure-sensitive adhesive layer 1 is provided only on one surface of the optical film 4. Fig. 5 shows a case where the second surface f2 of the pressure-sensitive adhesive layer 1 has the support 3b.

[0100]

<Optical film>

As the optical film, for example, a material used for forming an image display device such as a liquid crystal display or the like is used, and its type is not particularly limited. For example, the optical film may be a polarizing film. The polarizing film having a transparent protective film on one surface or both surfaces of a polarizer is generally used.

[0101]

<Polarizing Film>

The above-mentioned polarizing film may be a polarizing film having a polarizer, and a transparent protective film on at least one surface of the polarizer.

[0102]

The polarizer is not particularly limited, and may be a polarizer of various types. For example, the polarizer may be a product produced by a process including adsorbing a dichroic material such as iodine or a dichroic dye to a hydrophilic polymer film such as a polyvinyl alcohol-based film, a partially-formalized polyvinyl alcohol-based film, or a partially-saponified, ethylene-vinyl acetate copolymer-based film and uniaxially stretching the film, or may be a polyene-based oriented film such as a film of a dehydration product of polyvinyl alcohol or a

dehydrochlorination product of polyvinyl chloride. Among these polarizers, a polarizer including a polyvinyl alcohol-based film and a dichroic material such as iodine is preferred. The thickness of these polarizers is not particularly limited, and is generally from about 5 to 80  $\mu\text{m}$ .

[0103]

The polarizer yielded by dyeing a polyvinyl alcohol-based film with iodine, and then stretching the resultant uniaxially may be produced, for example, by immersing a polyvinyl alcohol into an aqueous solution of iodine to dye the polyvinyl alcohol, and then stretching the resultant into a length 3 to 7 times the original length. If necessary, the polyvinyl alcohol may also be immersed in an aqueous solution of potassium iodide or the like optionally containing boric acid, zinc sulfate, zinc chloride, or other materials. If necessary, the polyvinyl alcohol-based film may be further immersed in water for washing before it is dyed. If the polyvinyl alcohol-based film is washed with water, dirt and any anti-blocking agent can be cleaned from the surface of the polyvinyl alcohol-based film, and the polyvinyl alcohol-based film can also be allowed to swell so that unevenness such as uneven dyeing can be effectively prevented. The film may be stretched before, while, or after it is dyed with iodine. The film may also

be stretched in an aqueous solution of boric acid, potassium iodide, or the like or in a water bath.

[0104]

In the present invention, a thin polarizer of 10  $\mu\text{m}$  or less thickness may be used. For making the polarizing film thinner, the thickness is preferably from 1 to 7  $\mu\text{m}$ . Such a thin polarizer is preferred in the point that the polarizer is small in thickness unevenness, excellent in visibility, and small in dimension change to be excellent in endurance and further the thickness of the polarizing film is also made smaller.

[0105]

Typical examples of the thin polarizer include thin polarizing membranes described in JP-A-S51-069644, JP-A-2000-338329, WO2010/100917 pamphlet, JP-B1-4751481, and JP-A-2012-73563. These thin polarizing membranes can each be yielded by a producing method including the step of stretching a polyvinyl alcohol-based resin (hereinafter referred to as a PVA-based resin) and a resin substrate for stretching in a laminated state, and the step of dyeing the laminate. This producing method makes it possible to stretch the laminate, even when PVA-based resin layer is thin, without causing any inconvenience, such as breaking, by the stretching based on the supporting of the PVA-based resin layer on the resin substrate for stretching.

[0106]

The thin polarizing membranes are preferably polarizing membranes each yielded by the following producing method, among producing methods including the step of stretching the members concerned in the laminate state and the step of dyeing the laminate, since the laminate can be stretched into a large stretch ratio to improve the resultant in polarizing performance: a producing method including the step of stretching the laminating in an aqueous solution of boric acid, as is described in WO 2010/100917 pamphlet, JP-B1-4751481, or JP-A-2012-073563; and the membranes are in particular preferably membranes each yielded by a producing method including the step of stretching the laminate supplementally in the air before the stretching in the aqueous solution of boric acid, as is described in JP-B1-4751481, or JP-A-2012-073563.

[0107]

A thermoplastic resin with a high level of transparency, mechanical strength, thermal stability, moisture blocking properties, isotropy, and the like may be used as a material for forming a transparent protective film. Specific examples of such a thermoplastic resin include cellulose resins such as triacetylcellulose, polyester resins, polyethersulfone resins, polysulfone

resins, polycarbonate resins, polyamide resins, polyimide resins, polyolefin resins, (meth)acrylic resins, cyclic polyolefin resins (norbornene-based resins), polyarylate resins, polystyrene resins, polyvinyl alcohol resins, and a mixture thereof. The transparent protective film may be bonded with a pressure-sensitive adhesive layer to one side of the polarizer. On the other side of the polarizer, a thermosetting or ultraviolet-curable resin such as a (meth)acrylic, urethane, acrylic urethane, epoxy, and silicone resin may be used as the transparent protective film. The transparent protective film may contain any one or more suitable additives. Such additives include, for example, ultraviolet absorbers, antioxidants, lubricants, plasticizers, release agents, anti-coloring agents, flame retardants, nucleating agents, antistatic agents, pigments, and colorants. The content of the thermoplastic resin in the transparent protective film is preferably 50 to 100% by weight, more preferably 50 to 99% by weight, still more preferably 60 to 98% by weight, and particularly preferably 70 to 97% by weight. If the content of the thermoplastic resin in the transparent protective film is 50% by weight or less, high transparency and other properties inherent in the thermoplastic resin may be insufficiently exhibited.

[0108]

The thickness of the transparent protective film may

be appropriately determined, and is generally from about 1 to 500  $\mu\text{m}$  from the viewpoint of the strength, the handleability and other workabilities, the thin film property, and other properties of the film.

[0109]

On a surface of the transparent protective film to which the polarizer is not bonded, a functional layer such as a hard coat layer, an anti-reflection layer, or an anti-sticking layer can be formed, and treatment for the purpose of diffusion or anti-glare may be applied.

[0110]

The adhesive used to bond the polarizer to the transparent protective film may be any of various optically-transparent adhesives, such as aqueous adhesives, solvent type adhesives, hot melt type adhesives, radical-curable type adhesives, and cationically curable type adhesives, among which aqueous adhesives or radical-curable type adhesives are preferred.

[0111]

Examples of the optical film include a reflector, an anti-transmission plate, a retardation film (including a wavelength plate such as a half or quarter wavelength plate), a viewing angle compensation film, a brightness enhancement film, and any other optical layer that can be used to form liquid crystal displays or other devices.

They may be used alone as the optical film, or one or two or more layers of any of them may be used together with the polarizing film to form a laminate for practical use. A retardation film can also be used as the transparent protective film. As the retardation film, one obtained by stretching and/or contracting a polymer film or one obtained by aligning and fixing a liquid crystal material can be appropriately used according to the purpose.

[0112]

The optical film including a laminate of the polarizing film and the optical layer may be formed by a method of laminating them one by one in the process of manufacturing a liquid crystal display or the like. However, an optical film formed in advance by lamination is advantageous in that it can facilitate the process of manufacturing a liquid crystal display or the like because it has stable quality and good assembling workability. Proper adhesion means, such as a pressure-sensitive adhesive layer, may be used for laminating. On the occasion of adhesion of the aforementioned polarizing film and other optical layers, the optical axis may be set as a suitable configuration angle according to the target retardation characteristics, etc.

[0113]

An optical laminate B of the present invention has

the pressure-sensitive adhesive layer-attached optical film A and a low-refractive-index optical member 5 bonded to the pressure-sensitive adhesive layer 1 of the pressure-sensitive adhesive layer-attached optical film A. The optical member 5 is provided on the second surface f2 side of the pressure-sensitive adhesive layer 1. There is exemplified a case where in the optical laminate B shown in Fig. 6, the optical member 5 is bonded to the pressure-sensitive adhesive layer 1 after the support 3b (for example, a release film) is peeled off from the pressure-sensitive adhesive layer-attached optical film A shown in Fig. 5. Examples of the optical member 5 include an anti-reflection film, a light diffusion film, a prism film, a light guide film, a lens film, a Fresnel lens, a lenticular lens, and a microlens film.

[0114]

The pressure-sensitive adhesive layer-attached optical film or the optical laminate of the present invention can be preferably used for forming various image display devices such as liquid crystal displays. Liquid crystal displays may be formed according to conventional techniques. That is, a liquid crystal display may be typically formed by appropriately assembling a display panel such as a liquid crystal cell, a pressure-sensitive adhesive layer-attached optical film or an optical

laminate, and a component such as a lighting system as needed, and incorporating a driving circuit according to any conventional techniques, as long as the pressure-sensitive adhesive layer-attached optical film or the optical laminate according to the present invention is used. Any type of liquid crystal cell may also be used such as a TN type, an STN type, a  $\pi$  type a VA type and IPS type.

[0115]

An appropriate liquid crystal display such as a liquid crystal display in which a pressure-sensitive adhesive layer-attached optical film or an optical laminate is disposed on one side or both sides of a display panel such as a liquid crystal cell, or a liquid crystal display using a backlight or a reflector in a lighting system can be formed. In this case, the pressure-sensitive adhesive layer-attached optical film or the optical laminate according to the present invention may be installed in one side or both sides of a display panel of the liquid crystal cell or the like. When the optical films are provided in both sides, they may be of the same type or of different type. Furthermore, in assembling a liquid crystal display, suitable parts, such as diffusion plate, anti-glare layer, anti-reflection film, protective plate, prism array, lens array sheet, optical diffusion plate, and backlight, may be

installed in suitable position in one layer or two or more layers.

#### EXAMPLE

[0116]

The present invention is more specifically described by Examples below, which are not intended to limit the scope of the invention. In each Example, parts and percentages are all on a weight basis. Unless otherwise stated below, the conditions of room temperature standing are 23°C and 65% RH in all the cases.

[0117]

(Release film)

A 38  $\mu$ m-thick polyester film (trade name: DIAFOIL MRF, manufactured by Mitsubishi Plastics, Inc.) having one surface subjected to a release treatment using silicone was used.

[0118]

Comparative Example 1

(Preparation of pressure-sensitive adhesive composition (A))

41 parts by weight of 2-ethylhexyl acrylate (2EHA), 41 parts by weight of isostearyl acrylate (ISTA), 14 parts by weight of N-vinyl-2-pyrrolidone (NVP), 4 parts by weight of N-2-hydroxybutyl acrylate (4HBA), and two types of

photopolymerization initiators: 0.035 parts by weight of a photopolymerization initiator (trade name: IRGACURE 184, manufactured by BASF SE.) and 0.035 parts by weight of a photopolymerization initiator (trade name: IRGACURE 651, manufactured by BASF SE.) were put into a four-neck flask to prepare a monomer mixture. Subsequently, this monomer mixture was subjected to exposure of ultraviolet light in a nitrogen atmosphere in such a manner as to be partially photo-polymerized, thereby obtaining a partially polymerized product (acrylic-based polymer syrup) having a polymerization rate of about 10% by weight. 0.025 parts by weight of trimethylolpropane triacrylate (TMPTA) and 0.3 parts by weight of a silane coupling agent (trade name: KBM-403, manufactured by Shin-Etsu Chemical Co., Ltd.) were added to 100 parts by weight of the acrylic-based polymer syrup obtained as described above, and then uniformly mixed together to prepare a pressure-sensitive adhesive composition (A).

[0119]

(Production of pressure-sensitive adhesive layer (A))

The pressure-sensitive adhesive composition (A) was applied on a release-treated surface of the release film in such a manner that the thickness after forming the pressure-sensitive adhesive layer was 100  $\mu\text{m}$  to form an applied layer. Then, another release film was covered onto

a surface of the applied layer in such a manner that a release-treated surface faced the applied layer. Subsequently, the applied layer was photocured by irradiation with ultraviolet light under conditions including an illuminance of 6.5 mW/cm<sup>2</sup>, a light intensity of 2,000 mJ/cm<sup>2</sup>, and a peak wavelength of 350 nm to form a pressure-sensitive adhesive layer (A), so that a pressure-sensitive adhesive sheet (substrate-less type, thickness of the pressure-sensitive adhesive layer: 100 μm) provided with the release films on both surfaces of the pressure-sensitive adhesive layer (A) was prepared. A refractive index value ( $n_D$ ) of a D-line of the pressure-sensitive adhesive layer (A) measured by an Abbe refractometer at 23°C was 1.48, and the gel fraction was 67%.

[0120]

#### Comparative Example 2

(Preparation of pressure-sensitive adhesive composition (B))

To a monomer mixture consisting of 76 parts by weight of 2-ethylhexyl acrylate (2EHA), 18 parts by weight of N-vinyl-2-pyrrolidone (NVP), and 16 parts by weight of 2-hydroxyethyl acrylate (HEA), 0.050 parts by weight of 1-hydroxycyclohexyl phenyl ketone (trade name: IRGACURE 184, manufactured by BASF SE., having a light absorption band in a wavelength range of 200 to 370 nm) and 0.050 parts by

weight of 2,2-dimethoxy-1,2-diphenylethan-1-one (trade name: IRGACURE 651, manufactured by BASF SE., having a light absorption band in a wavelength range of 200 to 380 nm) were added as photopolymerization initiators. Then, the resulting mixture was irradiated with ultraviolet light until a viscosity thereof reached about 20 Pa·s (measurement conditions: BH viscometer, No. 5 rotor, 10 rpm, measurement temperature 30°C) to obtain a prepolymer composition (polymerization rate: 9%) in which the above monomer composition was partially polymerized. Then, 0.060 parts by weight of hexanediol diacrylate (HDDA) and 0.3 parts by weight of a silane coupling agent (trade name: KBM-403, manufactured by Shin-Etsu Chemical Co., Ltd.) were added to and mixed with the prepolymer composition to obtain a pressure-sensitive adhesive composition (b).

[0121]

(Preparation of pressure-sensitive adhesive composition (B))

With respect to the pressure-sensitive adhesive composition (b) obtained above (monomer component forming acrylic-based polymer was 100 parts by weight), 0.8 parts by weight (solid content weight) of 2,4-bis-[[4-(4-ethylhexyloxy)-4-hydroxy}-phenyl]-6-(4-methoxyphenyl)-1,3,5-triazine (trade name: Tinosorb S, manufactured by BASF Japan Ltd., absorption maximum wavelength of

absorption spectrum: 346 nm) dissolved in butyl acrylate in such a manner that the solid content was 15% and 0.3 parts by weight of bis(2,4,6-trimethylbenzoyl)-phenylphosphine oxide (trade name: IRGACURE 819, manufactured by BASF SE., having a light absorption band in a wavelength range of 200 to 450 nm) were added and stirred to obtain a pressure-sensitive adhesive composition (B).

[0122]

(Production of pressure-sensitive adhesive layer (B))

The pressure-sensitive adhesive composition (B) was applied on a release-treated surface of the release film in such a manner that the thickness after forming the pressure-sensitive adhesive layer was 150  $\mu\text{m}$  to form an applied layer. Then, another release film was covered onto a surface of the applied layer in such a manner that a release-treated surface faced the applied layer. Subsequently, the applied layer was photocured by irradiation with ultraviolet light under conditions including an illuminance of 6.5  $\text{mW}/\text{cm}^2$ , a light intensity of 2,000  $\text{mJ}/\text{cm}^2$ , and a peak wavelength of 350 nm to form a pressure-sensitive adhesive layer (B), so that a pressure-sensitive adhesive sheet (substrate-less type, thickness of the pressure-sensitive adhesive layer: 150  $\mu\text{m}$ ) provided with the release films on both surfaces of the pressure-sensitive adhesive layer (B) was prepared. The refractive

index value ( $n_D$ ) of the D-line of the pressure-sensitive adhesive layer (B) measured by an Abbe refractometer at 23°C was 1.49, and the gel fraction was 88%.

[0123]

### Comparative Example 3

(Preparation of pressure-sensitive adhesive composition (C))

95 parts of weight of butylacrylate (BA), 5 parts by weight of acrylic acid (AA), 0.2 parts by weight of azobisisobutyronitrile as a polymerization initiator, and 233 parts by weight of ethyl acetate were put into a separable flask provided with a thermometer, a stirrer, a reflux cooling tube and a nitrogen introducing tube, and a nitrogen gas was then fed to perform nitrogen purge for about 1 hour while the mixture was stirred. Thereafter, the flask was heated to 60°C, and the mixture was reacted for 7 hours to obtain an acrylic-based polymer having a weight average molecular weight ( $M_w$ ) of 1.1 million. To the acrylic-based polymer solution (the solid content was 100 parts by weight), 0.8 parts by weight of trimethylolpropane tolylene diisocyanate (trade name: Coronate L, manufactured by Nippon Polyurethane Industry Co., Ltd.) as an isocyanate-based cross-linking agent and 0.1 parts by weight of a silane coupling agent (trade name: KBM-403, manufactured by Shin-Etsu Chemical Co., Ltd.) were

added to prepare a pressure-sensitive adhesive composition (C: solution).

[0124]

(Production of pressure-sensitive adhesive layer (C))

The pressure-sensitive adhesive composition (C: solution) was applied on the release-treated surface of the release film in such a manner that the thickness after drying was 23  $\mu\text{m}$ , and then dried at 100°C for 3 minutes to remove the solvent, thus obtaining a pressure-sensitive adhesive layer (C). Thereafter, the pressure-sensitive adhesive layer was heated at 50°C for 48 hours to perform a crosslinking treatment. On an exposed surface of the obtained pressure-sensitive adhesive layer (C), another release film was covered in such a manner that the release-treated surface faced the exposed surface, so that a pressure-sensitive adhesive sheet (substrate-less type, thickness of the pressure-sensitive adhesive layer: 23  $\mu\text{m}$ ) provided with the release films on both surfaces of the pressure-sensitive adhesive layer (C) was prepared. The refractive index value ( $n_D$ ) of the D-line of the pressure-sensitive adhesive layer (C) measured by an Abbe refractometer at 23°C was 1.47, and the gel fraction was 82%.

[0125]

Comparative Example 4

(Preparation of pressure-sensitive adhesive composition (D))

99 parts by weight of butylacrylate (BA), and 1 part by weight of 4-hydroxybutylacrylate (4HBA) as monomer components, 0.2 parts by weight of azobisisobutyronitrile as a polymerization initiator, and ethyl acetate as a polymerization medium were put in such amounts that a solid content became 30% by weight into a separable flask provided with a thermometer, a stirrer, a reflux cooling tube and a nitrogen introducing tube, and a nitrogen gas was then fed to perform nitrogen purge for about 1 hour while the mixture was stirred. Thereafter, the flask was heated to 60°C, and the mixture was reacted for 7 hours to obtain an acrylic-based polymer having a weight average molecular weight (Mw) of 1.1 million. 0.11 parts of trimethylolpropane-xylylene diisocyanate ("Takenate D110N" manufactured by Mitsui Chemicals, Inc.) as an isocyanate-based cross-linking agent, and 0.1 parts of a silane coupling agent ("KBM-403" manufactured by Shin-Etsu Chemical Co., Ltd.) were added to the above acrylic-based polymer solution (solid content: 100 parts) to prepare a pressure-sensitive adhesive composition (D: solution).

[0126]

(Production of pressure-sensitive adhesive layer (D))

The pressure-sensitive adhesive composition (D:

solution) was applied on the release-treated surface of the release film in such a manner that the thickness after drying was 20  $\mu\text{m}$ , and then dried at 120°C for 3 minutes to remove the solvent, thus obtaining a pressure-sensitive adhesive layer (D). Thereafter, the pressure-sensitive adhesive layer was heated at 50°C for 48 hours to perform a crosslinking treatment. On an exposed surface of the obtained pressure-sensitive adhesive layer (D), another release film was covered in such a manner that the release-treated surface faced the exposed surface, so that a pressure-sensitive adhesive sheet (substrate-less type, thickness of the pressure-sensitive adhesive layer: 23  $\mu\text{m}$ ) provided with the release films on both surfaces of the pressure-sensitive adhesive layer (D) was prepared. The refractive index value ( $n_D$ ) of the D-line of the pressure-sensitive adhesive layer (D) measured by an Abbe refractometer at 23°C was 1.47, and the gel fraction was 75%.

[0127]

#### Example 1

(Preparation of dispersion containing low refractive index particles)

As a dispersion containing low refractive index particles, hollow nanosilica particles (hollow particles, refractive index: 1.24, average primary particle diameter:

75 nm, trade name: Throughrear 5320, manufactured by JGC Catalysts and Chemicals Ltd.) was diluted with a dispersion medium (methyl ethyl ketone/methyl isobutyl ketone=9/1:volume ratio) to prepare a dispersion having a particle concentration of 1.5% by weight.

[0128]

(Production of pressure-sensitive adhesive layer with adjusted refractive index)

One of the release films of the pressure-sensitive adhesive sheet obtained in Comparative Example 1 was peeled off. The above-described dispersion was applied to the surface of the exposed pressure-sensitive adhesive layer (A) by using a bar coater RDS No. 3 in such a manner that a refractive index adjustment region had a thickness of about 20 nm to 150 nm, and then dried in a drying oven at 110°C for 180 seconds. Then, a second release film was newly bonded to the surface of the pressure-sensitive adhesive layer (A) from which the hollow nanosilica particles were dispersed, to obtain a pressure-sensitive adhesive sheet. The release film on an opposite side of the second release film is referred to as the first release film.

[0129]

Examples 2 to 8

A pressure-sensitive adhesive sheet having a pressure-sensitive adhesive layer with an adjusted

refractive index was prepared in the same manner as in Example 1, except that the type of pressure-sensitive adhesive layer and the type of dispersion (the type of low refractive index particles, its average particle diameter, the type of dispersion medium, the particle concentration) in Example 1 were changed as shown in Table 1.

When a target thickness of the refractive index adjustment region after drying was about over 150 to 300 nm, a bar coater RDS No. 5 was used. When the target thickness of the refractive index adjustment region after drying was about 20 to 150 nm, a bar coater RDS No. 3 was used.

[0130]

(Evaluation)

The pressure-sensitive adhesive layers (pressure-sensitive adhesive sheets) obtained in Examples and Comparative Examples were evaluated as described below, and Table 1 shows the evaluation results.

[0131]

<Measurement of average surface refractive index>

An average surface refractive index of the pressure-sensitive adhesive layer (refractive index adjustment region side: second surface) obtained in Examples was calculated by measuring refractive indexes in the sodium D-lines (589 nm) using a spectroscopic ellipsometer (EC-400

manufactured by JA. Woolam Co.). From the pressure-sensitive adhesive sheets obtained in Examples and Comparative Examples, the release films on both surfaces of the pressure-sensitive adhesive sheet were peeled off, and while a black plate was bonded to the surface (first surface) not coated with the dispersion, an average refractive index of the surface (second surface) coated with the dispersion was measured. In the pressure-sensitive adhesive sheets of Comparative Examples, both the release sheets were peeled off, and then a black plate was bonded to one of the surfaces. In this state, an average refractive index of the pressure-sensitive adhesive layer surface was measured. In the pressure-sensitive adhesive layers of the pressure-sensitive adhesive sheets of Comparative Examples, the refractive indexes of both surfaces are the same.

[0132]

<Measurement of thickness of refractive index adjustment region>

A cross-section of the pressure-sensitive adhesive layer in a depth direction was adjusted to perform TEM observation. Based on the resulting TEM image (direct magnification: 3,000 to 30,000 times), a thickness of the refractive index adjustment region was measured. The thickness of the refractive index adjustment region was

determined as an average value of undulation of an interface between the region where the particles in the pressure-sensitive adhesive layer were dispersed and the region where no particles were dispersed. When it was difficult to identify the interface, an interfacial TEM image was subjected to binary image processing using image processing software (Image J), and a depth in a region where 90% (area) of the particles existed was determined as the thickness.

[0133]

<Total light transmittance and haze>

The second release film (second surface side of the pressure-sensitive adhesive layer) was peeled off from the pressure-sensitive adhesive sheet obtained in Example, and the resulting exposed surface was bonded to a glass slide (trade name: ShiroKenma No. 1, manufactured by Matsunami Glass Ind., Ltd., thickness: 0.8 to 1.0 mm, total light transmittance: 92%, haze: 0.2%). The other first release film was further peeled off to produce a test piece having a layer structure of the pressure-sensitive adhesive layer (the refractive index adjustment region was located on the glass slide side) and the glass slide. On the other hand, in the pressure-sensitive adhesive layer in Comparative Examples, one of the release films was peeled off, and the resulting exposed surface was bonded to a glass slide

similar to the above. The other release film was peeled off to prepare a test piece having a layer structure of the pressure-sensitive adhesive layer and the glass slide. For each of the test pieces, a total light transmittance and a haze value in a visual light range were measured using a haze meter (device name: HM-150, manufactured by Murakami Color Research Laboratory Co., Ltd).

[0134]

<Adhesiveness>

Sheet pieces having a length of 100 mm and a width of 20 mm were cut out from the pressure-sensitive adhesive sheets obtained in Examples and Comparative Examples. Then, the first release film (the side where no dispersion was applied in the pressure-sensitive adhesive layer) was peeled off from the sheet piece obtained from the pressure-sensitive adhesive sheet of Example, and then a PET film (trade name: LUMIRROR S-10, manufactured by TORAY Industries Inc., thickness: 25  $\mu$ m) was attached (lined) to the pressure-sensitive adhesive layer surface. Subsequently, the second release film was peeled off, and then the sheet piece was press-bonded to a glass plate (trade name: Soda-Lime Glass #0050, manufactured by Matsunami Glass Ind., Ltd.) as a test plate, under press-bonding conditions: 2 kg roller; and one stroke, to prepare a sample constituted of the test plate/the pressure-

sensitive adhesive layer (the first surface was located on the PET side)/the PET film. On the other hand, in the sheet piece obtained from the pressure-sensitive adhesive sheet of Comparative Example, after one of the release films was peeled off, a PET film similar to the above was bonded to the pressure-sensitive adhesive layer surface. Then, the other release film was peeled off to prepare a sample using a test plate similar to the above. Each of the obtained samples was subjected to an autoclave treatment (50°C, 0.5 MPa, 15 minutes), and then subjected to cooling in an atmosphere at 23°C and 50% R.H., for 30 minutes. After the cooling, the pressure-sensitive adhesive sheet (the pressure-sensitive adhesive layer/the PET film) was peeled off from the test plate to measure an adhesive strength (N/20 mm) during 180-degree peeling, using a tension tester (device name: Autograph AG-IS, manufactured by Shimadzu Corp.) according to JIS Z0237, in an atmosphere at 23°C and 50% R.H., under conditions including a tension rate of 300 mm/minute and a peeling angle of 180 degrees.

[0135]

#### <Measurement of surface reflectance>

The surface (second surface) of the pressure-sensitive adhesive layer obtained in Example on the side coated with the dispersion was used as a reflectance

measuring surface. The first release film (the side where no dispersion was applied in the pressure-sensitive adhesive layer) was peeled off from the pressure-sensitive adhesive sheet obtained in Example, and a black acrylic plate (trade name "CLAREX", manufactured by Nitto Jushi Kogyo Co., Ltd.) was bonded. Then, the second release film (the side where the dispersion was applied in the pressure-sensitive adhesive layer) was peeled off, and the peeled surface was used as a sample for surface reflectance measurement. On the other hand, for the pressure-sensitive adhesive layer obtained in Comparative Example, after one of the release films was peeled off from the pressure-sensitive adhesive sheet, a black acrylic plate similar to the above was bonded thereto. Then, the other release film was peeled off, and the peeled surface was used as a sample for surface reflectance measurement. A surface reflectance (Y value) was measured by a spectrophotometer (U4100, manufactured by Hitachi High-Technologies Corporation).

[0136]

<Measurement of internal reflection suppression rate  
(effect of improving transmittance)>

After the second release film (the side where the dispersion was applied in the pressure-sensitive adhesive layer) was peeled off from the pressure-sensitive adhesive sheet obtained in Example, the pressure-sensitive adhesive

layer surface was bonded to a low refractive layer side of a laminated film in which a low refractive index layer having a refractive index of 1.36 was formed on a triacetyl cellulose film, and lamination was performed such that a low refractive index adjustment region of the pressure-sensitive adhesive layer was in contact with the low refractive index layer on the laminated film. Next, the first release film was peeled off, and a glass slide (trade name: ShiroKenma No. 1, manufactured by Matsunami Glass Ind., Ltd., thickness: 0.8 to 1.0 mm, total light transmittance: 92%, haze: 0.2%) was bonded to the pressure-sensitive adhesive layer surface. Thus, a test piece having a layer structure of the triacetyl cellulose film/ the low refractive index layer/ the pressure-sensitive adhesive layer (the second surface was located on the low refractive index layer side)/ the glass slide was prepared. On the other hand, for the pressure-sensitive adhesive layer obtained in Comparative Example, after one of the release films was peeled off from the pressure-sensitive adhesive sheet, in the same manner as above, a test piece having a layer structure of the triacetyl cellulose film/ the low refractive index layer/ the pressure-sensitive adhesive layer/ the glass slide was prepared.

[0137]

The internal reflection suppression rate (effect of

improving the transmittance) was calculated based on the following formula by measuring the transmittance of the test piece prepared above. In the following formula, "transmittance (%) in case devoid of the particles" means a reflectance of the test piece in Comparative Example. That is, the internal reflection suppression effect (effect of improving the transmittance) is an index indicating how much the internal reflectance can be reduced by providing a refractive index adjustment layer.

Internal reflection suppression rate (%) =  
"transmittance (%)" - "transmittance (%) in case devoid of the particles"

[0138]

[Table 1]

| PRESSURE-SENSITIVE ADHESIVE LAYER                  |   |      |                                      |                      |                                                    |                  |                                |                                                                  |              | DISPERSION                 |                              | EVALUATION |                                               |                                         |     |                                                                            |     |                          |
|----------------------------------------------------|---|------|--------------------------------------|----------------------|----------------------------------------------------|------------------|--------------------------------|------------------------------------------------------------------|--------------|----------------------------|------------------------------|------------|-----------------------------------------------|-----------------------------------------|-----|----------------------------------------------------------------------------|-----|--------------------------|
| BASE OF PRESSURE-SENSITIVE ADHESIVE LAYER (MATRIX) |   | TYPE | FIRST SURFACE: REFRACTIVE INDEX (n1) | THICKNESS (μm)       | DISPERSION REGION OF LOW REFRACTIVE INDEX MATERIAL |                  |                                |                                                                  | DISPERSION   | PARTICLE CONCENTRATION [%] | TRANSMISSION CHARACTERISTICS |            | 180-DEGREE PEELING ADHESIVE STRENGTH [N/20mm] | GLUE SURFACE REFLECTION CHARACTERISTICS |     | INTERNAL REFLECTION SUPPRESSION EFFECT (EFFECT OF IMPROVING TRANSMITTANCE) |     |                          |
|                                                    |   |      |                                      |                      | TYPE                                               | REFRACTIVE INDEX | AVERAGE PARTICLE DIAMETER [NM] | SECOND SURFACE: REFRACTIVE INDEX (n2) : AVERAGE REFRACTIVE INDEX |              |                            |                              |            |                                               |                                         |     |                                                                            |     | DEPTH T OF PARTICLE [mm] |
|                                                    |   |      |                                      |                      |                                                    |                  |                                |                                                                  |              |                            |                              |            |                                               |                                         |     |                                                                            |     |                          |
| EXAMPLE 1                                          | A | 1.48 | 100                                  | (X1) HOLLOW PARTICLE | 1.24                                               | 75               | 1.35                           | 140                                                              | MEK/MIBK     | 1.5                        | 91.5                         | 0.8        | 10.8                                          | 2.0                                     | 2.0 | 90.8                                                                       | 0.8 |                          |
| EXAMPLE 2                                          | A | 1.48 | 100                                  | (X1) HOLLOW PARTICLE | 1.24                                               | 75               | 1.36                           | 100                                                              | ETHANOL/MIBK | 1.4                        | 91.2                         | 0.6        | 12.2                                          | 1.6                                     | 2.4 | 91.0                                                                       | 1.0 |                          |
| EXAMPLE 3                                          | A | 1.48 | 100                                  | (X2) HOLLOW PARTICLE | 1.32                                               | 80               | 1.37                           | 100                                                              | IPA          | 1.5                        | 91.7                         | 0.8        | 9.2                                           | 2.4                                     | 1.6 | 90.5                                                                       | 0.5 |                          |
| EXAMPLE 4                                          | A | 1.48 | 100                                  | (X3) HOLLOW PARTICLE | 1.33                                               | 60               | 1.38                           | 150                                                              | IPA          | 2.0                        | 91.8                         | 0.6        | 11.8                                          | 2.4                                     | 1.6 | 90.5                                                                       | 0.5 |                          |
| EXAMPLE 5                                          | A | 1.48 | 100                                  | (Y1)MgF2             | 1.38                                               | 40               | 1.45                           | 80                                                               | ETHANOL      | 1.8                        | 92.2                         | 0.3        | 13.5                                          | 3.4                                     | 0.6 | 90.5                                                                       | 0.2 |                          |
| EXAMPLE 6                                          | C | 1.47 | 23                                   | (Y1)MgF2             | 1.38                                               | 40               | 1.44                           | 200                                                              | ETHANOL      | 2.0                        | 92.2                         | 0.5        | 8.8                                           | 3.3                                     | 0.7 | 90.5                                                                       | 0.2 |                          |
| EXAMPLE 7                                          | B | 1.49 | 150                                  | (Y2) CRYOLITE        | 1.34                                               | 50               | 1.41                           | 150                                                              | ETHANOL      | 2.0                        | 92.1                         | 0.5        | 12.5                                          | 3.0                                     | 1.0 | 90.4                                                                       | 0.4 |                          |
| EXAMPLE 8                                          | D | 1.47 | 20                                   | (Y2) CRYOLITE        | 1.34                                               | 50               | 1.42                           | 200                                                              | ETHANOL      | 2.0                        | 92.2                         | 0.4        | 2.3                                           | 3.2                                     | 0.8 | 90.3                                                                       | 0.3 |                          |
| COMPARATIVE EXAMPLE 1                              | A | 1.48 | 100                                  | -                    | -                                                  | -                | 1.48                           | -                                                                | -            | -                          | 92.2                         | 0.3        | 18.5                                          | 4.0                                     | 0.0 | 90.0                                                                       | -   |                          |
| COMPARATIVE EXAMPLE 2                              | B | 1.49 | 150                                  | -                    | -                                                  | -                | 1.49                           | -                                                                | -            | -                          | 92.3                         | 0.4        | 18.0                                          | 4.0                                     | 0.0 | 90.0                                                                       | -   |                          |
| COMPARATIVE EXAMPLE 3                              | C | 1.47 | 23                                   | -                    | -                                                  | -                | 1.47                           | -                                                                | -            | -                          | 92.3                         | 0.3        | 9.5                                           | 4.0                                     | 0.0 | 90.0                                                                       | -   |                          |
| COMPARATIVE EXAMPLE 4                              | D | 1.47 | 20                                   | -                    | -                                                  | -                | 1.47                           | -                                                                | -            | -                          | 92.4                         | 0.3        | 4.2                                           | 4.0                                     | 0.0 | 90.0                                                                       | -   |                          |

[0139]

Table 1 shows

(X1) hollow nanosilica particles: refractive index  
1.24 (trade name: Throughrear 5320, manufactured by JGC  
Catalysts and Chemicals Ltd., particle diameter: 75 nm),

(X2) hollow polymer fine particles: refractive index  
1.32 (trade name: Techpolymer NH, manufactured by Sekisui  
Kasei Co., Ltd., product number XX-255AA, particle diameter  
80 nm)

(X3) hollow polymer fine particles: refractive index  
1.33 (trade name: Techpolymer NH, manufactured by Sekisui  
Kasei Co., Ltd., product number XX-260AA, particle diameter  
60 nm)

(Y1)  $\text{MgF}_2$ : 1.38, average primary particle diameter:  
40 nm, manufactured by CIK-Nano Tek.,

(Y2)  $\text{Na}_3\text{AlF}_6$ : 1.34, average primary particle  
diameter: 50 nm, manufactured by CIK-Nano Tek.,  
MEK/MIBK methyl ethyl ketone/methyl isobutyl ketone = 9/1  
(volume ratio),

Ethanol/MIBK ethanol/methyl isobutyl ketone = 9/1  
(volume ratio), and

IPA isopropyl alcohol.

DESCRIPTION OF REFERENCE SIGNS

[0140]

1 PRESSURE-SENSITIVE ADHESIVE LAYER

1a BASE (MATRIX) OF WHOLE OF PRESSURE-SENSITIVE ADHESIVE  
LAYER

2 LOW REFRACTIVE INDEX MATERIAL

f1 FIRST SURFACE OF PRESSURE-SENSITIVE ADHESIVE LAYER

f2 SECOND SURFACE OF PRESSURE-SENSITIVE ADHESIVE LAYER

T THICKNESS OF REFRACTIVE INDEX ADJUSTMENT REGION

S SUPPORT

3a SUPPORT

3b SUPPORT

4 OPTICAL FILM

5 LOW REFRACTIVE OPTICAL MEMBER

10 DISPERSION

## CLAIMS

1. A pressure-sensitive adhesive layer comprising a first surface and a second surface on a side opposite to the first surface,

wherein the pressure-sensitive adhesive layer forms a base of a whole of the pressure-sensitive adhesive layer with a pressure-sensitive adhesive composition containing a base polymer, and

the first surface has a first refractive index based on the pressure-sensitive adhesive composition, while a second refractive index of the second surface is lower than the first refractive index of the first surface.

2. The pressure-sensitive adhesive layer according to claim 1 or 2, wherein a difference between the first refractive index of the first surface and the second refractive index of the second surface is 0.02 to 0.45.

3. The pressure-sensitive adhesive layer according to any one of claims 1 to 3, wherein the second refractive index of the second surface is 1.45 or less.

4. The pressure-sensitive adhesive layer according to any one of claims 1 to 3, wherein in the pressure-

sensitive adhesive layer, on a side of the second surface, a low refractive index material having a lower refractive index than the refractive index of the base polymer is dispersed.

5. The pressure-sensitive adhesive layer according to claim 4, wherein a thickness of a region where the low refractive index material is dispersed is 600 nm or less in a thickness direction from the side of the second surface in the pressure-sensitive adhesive layer.

6. The pressure-sensitive adhesive layer according to claim 4 or 5, wherein the base polymer has a refractive index of 1.40 to 1.55, and the low refractive material has a refractive index of 1.10 to 1.45.

7. The pressure-sensitive adhesive layer according to any one of claims 4 to 6, wherein a difference between the refractive index of the base polymer and the refractive index of the low refractive index material is 0.07 to 0.45.

8. The pressure-sensitive adhesive layer according to any one of claims 4 to 7, wherein the low refractive index material is particles having an average particle diameter of 10 nm to 150 nm.

9. The pressure-sensitive adhesive layer according to any one of claims 4 to 8, wherein the low refractive index material is at least one particle selected from a group consisting of at least one inorganic particle selected from a group consisting of  $\text{MgF}_2$ ,  $\text{CaF}_2$  and  $\text{Na}_3\text{AlF}_6$ , a porous silica particle, a hollow nanosilica particle, and a hollow polymer particle.

10. The pressure-sensitive adhesive layer according to any one of claims 1 to 9, wherein a total light transmittance is 85% or more.

11. The pressure-sensitive adhesive layer according to any one of claims 1 to 10, wherein the second surface has a reflectance of 0.5 to 3.5%.

12. The pressure-sensitive adhesive layer according to any one of claims 1 to 11, wherein a difference in reflectance between the first surface and the second surface is 0.1 to 3.5%.

13. The pressure-sensitive adhesive layer according to any one of claims 1 to 12, wherein a gel fraction is 30 to 95% by weight.

14. The pressure-sensitive adhesive layer according to any one of claims 1 to 13, wherein a storage elastic modulus  $G'$  at 25°C is 0.05 to 0.50 MPa.

15. The pressure-sensitive adhesive layer according to any one of claims 1 to 14, wherein a  $\tan \delta$  peak value is -5 to -50°C when a dynamic viscoelasticity is measured at 1 Hz.

16. A method for producing the pressure-sensitive adhesive layer according to any one of claims 1 to 15, comprising:

a step (1) of forming a base pressure-sensitive adhesive layer on a support with a pressure-sensitive adhesive composition containing a base polymer;

a step (2) of providing a dispersion in which a low refractive index material having a lower refractive index than the refractive index of the base polymer is dispersed;

a step (3) of applying the dispersion to a second surface on a side opposite to a first surface on a side of the support in the base pressure-sensitive adhesive layer and penetrating the low refractive index material contained in the dispersion in a thickness direction from the second surface in the base pressure-sensitive adhesive layer; and

a step (4) of drying the pressure-sensitive adhesive layer into which the low refractive index material has penetrated.

17. A pressure-sensitive adhesive sheet comprising: the pressure-sensitive adhesive layer according to any one of claims 1 to 15; and a support on one surface or both surfaces of the pressure-sensitive adhesive layer.

18. A pressure-sensitive adhesive layer-attached optical film comprising:

an optical film; and a pressure-sensitive adhesive layer provided on one surface or both surfaces of the optical film,

wherein the pressure-sensitive adhesive layer on the one surface or the both surfaces is the pressure-sensitive adhesive layer according to any one of claims 1 to 15, and the optical film is attached to the first surface side of the pressure-sensitive adhesive layer.

19. The pressure-sensitive adhesive layer-attached optical film according to claim 18, wherein the optical film is a polarizing film.

20. An optical laminate comprising: the pressure-

sensitive adhesive layer-attached optical film according to claim 18 or 19; and a low-refractive-index optical member bonded to the pressure-sensitive adhesive layer of the pressure-sensitive adhesive layer-attached optical film.

21. An image display device comprising the pressure-sensitive adhesive layer according to any one of claims 1 to 15, the pressure-sensitive adhesive layer-attached optical film according to claim 18 or 19, or the optical laminate according to claim 19.