PHYSICAL REVIEW B VOLUME 50, NUMBER 14 1 OCTOBER 1994-II

Experimental verification of light localization for disordered multilayers in the visible-infrared spectrum

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(Received 9 December 1993; revised manuscript received 8 April 1994)

We have measured the light localization length for a disordered binary multilayer system, which we have prepared by alternating layers of antimonite and cryolite. The thicknesses of the cryolite layers are all the same, while those of antimonite vary randomly around an average value. The degree of disorder is given by the average fluctuation of thicknesses. The exponential decay of transmission intensity of light with the length of the multilayer is demonstrated experimentally. The localization lengths, the characteristic length of this exponential decay, are derived from experimental data in the wavelength region 720–900 nm for a fixed degree of disorder. The localization lengths at different incident wavelengths are computed by transfer-matrix approach. The agreement between the experimental measurements and the calculations is good.

I. INTRODUCTION

Almost a decade ago, enhanced coherent backscattering, the precursor of light localization, was experimentally demonstrated, and named the weak-localization effect.[1–4] Since then, a good deal of both theoretical and experimental effort has been devoted to the investigation of light localization.[5–8] An important achievement was made by Genack and Garcia.[9] In a sample composed of a random mixture of aluminum and Teflon spheres they observed the exponential decay of the transmitted wave with increasing depth. This was attributed to Anderson localization of the microwaves, induced by the randomness. However, studies on photon elastic scattering show that it is difficult to meet the Ioffe-Regel criterion in the visible-infrared spectrum by increasing the density of random scatterers, and materials with higher dielectric constant are rare in nature. The barriers to getting strong light localization have led to an idea proposed by John:[10] the working medium may be dielectric structures which, although not statistically homogeneous, exhibit the statistical symmetry of a crystal. Monosized spheres with high dielectric constant held in a host medium form a Bragg-like diffraction grating; this kind of composite material is considered to be such a working medium, in which any small disorder can cause photon localization. This material is the optical analog of an amorphous semiconductor for electron localization. A disordered multilayer structure may be a suitable one-dimensional candidate for quantitatively examining effects due to the disorder of a periodic system.

It is known that a periodic binary multilayer possesses a band gap, which can be determined from the dispersion relation of light waves. Within the gap the transmission intensity of light varies exponentially with the length of the multilayer, while outside the gap this relationship fails. This phenomenon is caused physically by phase-coherent Bragg reflection. Theoretical studies have also shown that the exponential dependence always exists for a disordered one- dimensional system.[11] In other words, light can be localized in such a system. Kondilis and Tzanetakis[12] and McGurn *et al.*[13] dealt theoretically with light propagation in a one-dimensional disordered binary multilayer by the optical transfer-matrix method. It was confirmed that because of the disorder an exponential attenuation of transmission intensity versus depth held at all nonzero photon frequencies. Recently Frigerio, Rivory, and Sheng[14] reported simulated results for an experimentally achievable disordered one-dimensional system. By considering the density of states, they explained the dependence of localization length on the degree of disorder in the system in three distinguishable frequency regions, i.e., the photonic gap, the bandtail, and the passband. Nevertheless, as far as we know, there has not been experimental verification.

In this paper we report an experimental study of the changes in reflection and transmission as light passes through a disordered binary multilayer system which consists of two materials with high contrast of refractive index. When the layer thickness of one material is kept constant and that of the other material has a random distribution around an average value, the exponential relation between transmission and multilayer length holds for all detected wavelengths in the spectral region of 720–900 nm. By measuring transmission intensities the corresponding localization lengths are derived. Meanwhile, the transmission and the localization lengths of the binary multilayers are calculated in terms of the optical transfer-matrix approach. An excellent agreement between measured and calculated values is established. Finally, we briefly discuss the different effects caused by phase-coherent Bragg reflection and the disorder of the medium.

II. DISORDERED BINARY MULTILAYERS

Antimonite and cryolite are chosen as the coating materials because of their high contrast of refractive index

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and small absorption in the measuring range of 700–900 nm. Within this wavelength region their refractive indices are almost unchanged and are $n_b = 2.83$ and $n_a = 1.33$, respectively. This makes the refractive contrast as high as 2.1. A previous study has noted that higher refractive contrast may lead to a pronounced effect on the change of localization length.[12] In addition, their evaporation temperatures are rather low, so that using a simple method we can prepare homogeneous binary multilayers.

The disorder of the binary multilayer is produced by changing the thickness of the antimonite layers b_m, while that of all cryolite layers, a_m is the same. Quantitatively, $a_m = \langle a \rangle = d/4n_a = 122$ nm and b_m varies randomly around an average value $\langle b \rangle = d/4n_b = 57$ nm (here $d = 650$ nm). We use the average fluctuation of layer thickness, δb, to describe the disorder in the multilayers and $\delta b = [\sum_m (b_m - \langle b \rangle)^2/M]^{1/2}$, where b_m is the real thickness and M is the total number of antimonite layers. The average lattice constant is given by $l_{ab} = \langle a \rangle + \langle b \rangle$. The degree of disorder in the multilayer is then defined as $D = \delta b / l_{ab}$. To get a clear effect, a relatively large disorder $D = 0.16$ is used in this experiment. Three kinds of multilayer on glass substrates are prepared, with the same $D = 0.16$ and same average lattice constant l_{ab}, but different total lengths $L = 10$, 15, and 21 in units of l_{ab}, i.e., different numbers of periods.

The coating procedure is briefly described as follows. First the real thicknesses of the antimonite layers are decided based on the requirement that $D = 0.16$ in the cases of total lengths 10, 15, and 21. Then by using thermal evaporation the two materials are alternately coated. The thickness of the layer is monitored *in situ* by measuring the transmission of a probe light beam. The dependence of transmission of the probe beam on the monitored layer thickness is deduced from the standard formulation of thin-film optics. In order to have high monitoring sensitivity, the transmission intensity of the probe beam is recalibrated after completing each period of the film. The error of thickness can then be controlled to be within 2 nm. During the coating process the evaporation rate of the materials and the temperature of the substrate should be carefully adjusted, otherwise the tension existing in the film can induce many cracks. This results in an opaque film as the number of layers goes up.

A regular $d/4n$ binary film with $L = 10$ and made of the same materials is additionally prepared to test whether the control of the layer thickness works well. Figure 1 shows the comparison of its measured transmission spectrum with the theoretical one, which is calculated by using the designated thicknesses. The coincidence of the two curves is quite good, except in the region of wavelengths shorter than 600 nm. This is caused by the strong absorption of antimonite in this region, which makes the transmission drop seriously. The results shown in Fig. 1 mean that in our experiments the accuracy of the layer thickness is assured. Furthermore, in the measured spectrum there is a band gap located at wavelengths around 530–850 nm, which is also computed by means of the dispersion relation of incident light.[15] The corresponding dispersion relation for our regular film can be expressed as

$$2\pi d/\lambda = 4\arccos[0.435\cos(0.276dK)+0.565]^{1/2} .$$

In the first Brillouin zone, i.e., when the Bloch vector K varies from 0 to $\pi/(a+b)$, the gap should appear at wavelengths of 526–849 nm. The agreement between experiment and calculation is also established.

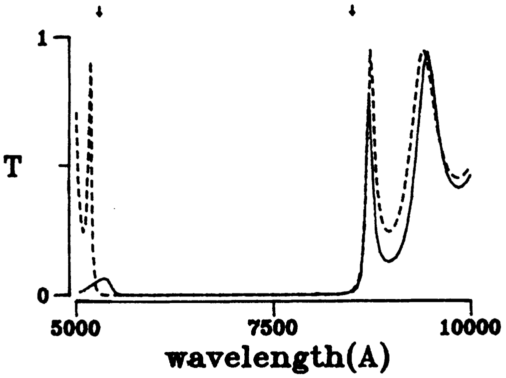

FIG. 1. Transmission spectra of regular $d/4n$ binary film with $d = 650$ nm and total length $L = 10$ (in units of the average lattice constant) between 500 and 1000 nm; solid line, measured; dashed line, calculated. Due to the absorption of antimonite the measured transmissions decrease at wavelengths shorter than 600 nm. A band gap around 650 nm is readily seen (between arrows).

III. MEASUREMENT OF TRANSMISSION AND REFLECTION OF DISORDERED MULTILAYERS

The effects of the disorder on light propagation are examined by measuring the reflection and transmission spectra, as light with wavelengths of 500–1100 nm travels through these films. Figure 2 shows the measured light transmission curve for a disordered multilayer with $L = 10, D = 0.16$ and a regular multilayer with $L = 10, D = 0$. Comparing these curves one can see an apparent difference, specifically inside the gap (526–849 nm). In the case of the ordered multilayer, the transmis-

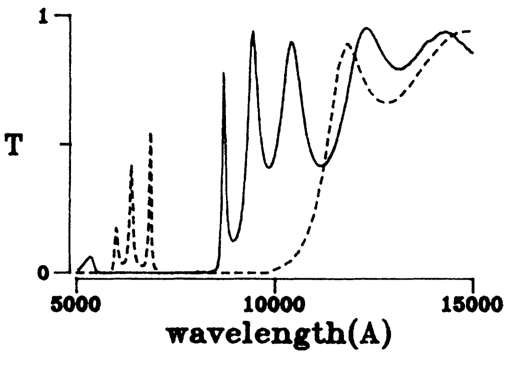

FIG. 2. Comparison of transmission spectra for regular and disordered multilayers of $L = 10$; solid curve, regular film; dashed curve, disordered film with degree of disorder $D = 0.16$. The total reflection inside the gap no longer exists for the disordered multilayer.

sion is almost zero in the whole gap, while for the disordered one there are three transmission peaks, located around 528, 584, and 654 nm and the maximum transmission can reach 85%. For thicker disordered films, $L=15$ and 21, those peaks still exist. There are small shifts of the peaks' wavelength, but rather big changes in transmission intensity. In the reflection spectrum decreases of reflectance at the noted wavelengths are observed too. The occurrence of these peaks may be attributed to the destruction of phase-coherent Bragg reflection caused by the disorder of the multilayer system.

IV. MEASUREMENT OF LOCALIZATION LENGTHS

The main part of the experiment is to get accurate transmission intensities near the gap edge. Then the experimental localization lengths are determined.

For a one-dimensional multilayer system it is more convenient to use the transfer-matrix approach to get the transmission and reflection of light. The transmission T and reflection R are connected directly with the elements of the transfer matrix M_{ij}. In the case of negligible absorption of the glass substrates used, R and T can be expressed by $R=[M_{21}/M_{11}]^2$ and $T=[1/M_{11}]^2$.

When light propagates through the multilayer, the localization length l_c is defined as $l_c=-L/\langle \ln T \rangle$. L is the total length of the multilayer in units of the average period l_{ab}; $\langle \cdots \rangle$ represents the average over the statistical distribution. It means that we should make the statistical average for varied multilayers which possess the same total length L and the same degree of disorder D. It is almost impossible to fabricate such a series of multilayers experimentally. Fortunately, the simulated results, plotted in Figs. 2 and 3 of Ref. 12, show that the fluctuation of l_c is not very serious, when the statistical average is not considered. So, in practice, for each L only a single layer-thickness distribution is chosen to measure and calculate T, and by using a linear regression for $T(L)$ the localization length l_c is derived. To compare with the measurements, in our simulation the number of layers that enters into the calculation is restricted to the real number, i.e., L is limited to 10, 15, and 21 in the present case. This is different from the methods used in Refs. 12 and 13, where the number of layers entering into the calculation is not limited. It might induce some errors, especially in the case of large l_c. However, our calculations verify that the error is small and acceptable so long as the ratio of transmission to incident intensity is lower than 10^{-3}. Physically, it means that the length of the multilayer should be much longer than the localization length. This requirement is well satisfied in the whole spectral range measured. According to our experiment, the absorption of antimonite should be taken into account when the wavelength of the incident light is shorter than 600 nm. To avoid the additional effect induced by the absorption, the working wavelength in our experiment is restricted to the region of 720–900 nm.

The light source used for getting accurate transmission intensity is a cw Ti: sapphire tunable laser pumped by an S-P model 171 Ar^+ laser. The output in the range 700–900 nm is around 500 mW with TEM_{00} mode and linear polarization. The diameter of the beam is about 1.5 mm with a very small divergence. The laser beam passes through two 2-mm-diam apertures, whose distance apart is about 30 cm. The functions of the apertures are to block stray radiation from the laser source and to assure that the laser beam is normally incident on the surface of the multilayer. Then the penetrating beam enters a box with a small window. In the box the multilayer and a highly sensitive photodiode are set. The detectable ratio of output to input may be better than 10^{-8}, which makes precise detection of very weak transmission possible. In the experiment great care must be taken to prevent all scattered and stray light coming from optical elements and holders, which can seriously disturb the results. For example, when the beam reflected from the surface of the multilayer does not pass inversely through the apertures, the scattered light from the edge of the aperture may be stronger than the transmission. The measurements are carried out at 11 wavelengths between

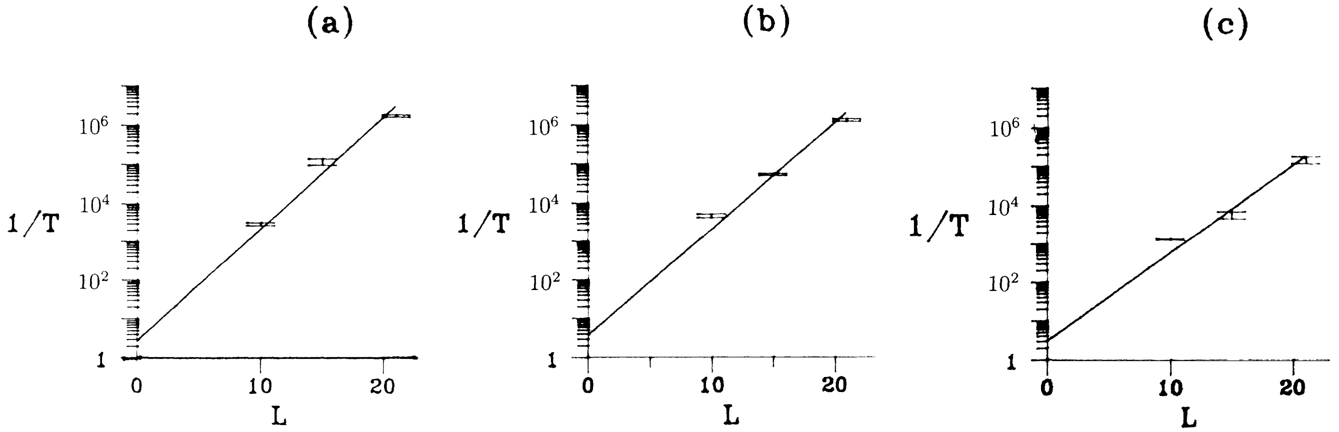

FIG. 3. Exponential dependence of reciprocal transmission $1/T$ on the length L of the multilayer at three incident wavelengths: (a) 734 nm, within the gap; (b) 830 nm, inside the gap edge; (c) 860 nm, outside the gap edge. The degrees of disorder of the measured multilayers are identical, $D=0.16$. The localization length l_c is decided by linear regression to be $1.43l_{ab}$, $1.49l_{ab}$, and $1.79l_{ab}$. The error bar represents the standard error of measured values that are detected at different positions of a film.

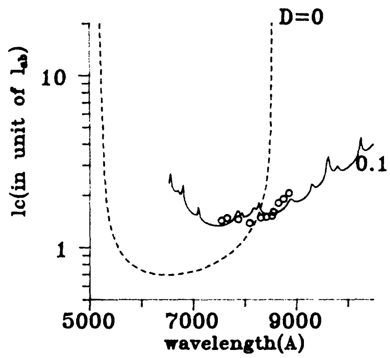

FIG. 4. Variation of localization length l_c in units of l_{ab} with incident wavelength λ between 700 and 1000 nm. *o*, experimental values; solid line, numerical simulation. To see the change of l_c in the gap edge the localization lengths in the corresponding regular film are plotted as the dashed line.

720 and 900 nm with three kinds of multilayers, which have the same $D=0.16$ but different total lengths $L=10$, 15, and 21. To get reliable transmission values and eliminate possible errors caused by inhomogeneity of layer thickness, the same measurement is repeated by changing the irradiation position on the multilayer. We arbitrarily choose at least nine small areas for each multilayer at each wavelength. The average value is taken as the final transmission intensity of the multilayer at the given wavelength. It is found that for different areas of a multilayer the change of transmission is rather small (shown as the error bar in Fig. 3). Therefore the variation of transmission intensity T with L is obtained, and from the plot of T^{-1}-L the localization length l_c can be determined by linear regression. Figure 3 shows this procedure at three measured wavelengths. The exponential relation between T^{-1} and L is obviously seen. This relationship is found to be kept for all detected wavelengths whether they are inside or outside the gap. It indicates that now, due to disorder, the localization effect exists not only within the gap but also outside the gap. The dependence of l_c on the wavelength of the incident light is given in Fig. 4. It is seen that near the gap edge the corresponding localization lengths are as short as around $1.5l_{ab}$. The experimental result agrees very well with the simulated curve both inside and outside the gap. Based on Figs. 3 and 4 we can confirm experimentally that disorder can really cause light localization in a one-dimensional multilayer.

In addition, to have a better understanding of the mechanism of light localization in a layered system the variation of localization length with wavelength at different degrees of disorder D is computed. The results are the same as those stated in Refs. 12–14: inside the gap a larger D produces a longer l_c, but outside the gap the opposite occurs. This phenomenon might be explained by the competition of two physical processes which can induce exponential attenuation of light versus depth. Inside the gap for an ordered multilayer, i.e., $D=0$, perfect phase-coherent Bragg reflection causes exponential decay of the light intensity, so that the transmission vanishes in a very short distance. This leads to a small l_c. Once the multilayer system is not longer ordered, for example, in the case of varying thicknesses, the coherence of reflected beams coming from the interfaces between two coating materials may be disturbed. The interference effect, i.e., Bragg reflection, will be diminished and Anderson localization induced by the disorder may start to play a role. If the degree of disorder is small, both processes should be in action simultaneously. However, as the disorder becomes more pronounced the influence of Bragg interference on l_c will become less and less; this makes l_c smooth and without any abrupt jump at the gap edge. This phenomenon has appeared in our experiment. In Fig. 4 a continuous and slow variation of l_c with the wavelength in the region of the gap edge might be evidence. It implies that in the case of a layered system with $D=0.16$ the disorder may be the dominant cause of localization. The situation outside the gap seems to be simple, because the disorder is the only reason for localization. A greater degree of disorder must correspond to a smaller localization length. The related experiments are in progress.

V. CONCLUSION

We have measured the light transmission for one-dimensional disordered binary multilayers, which are periodic on average. It is verified experimentally that incident light with a wavelength in the visible-infrared region can be localized. The localization length can become shorter than two average lattice constants near the gap edge. The agreement between the experiment and a numerical simulation performed by the transfer-matrix method is good.

ACKNOWLEDGMENTS

The authors wish to thank NSFC for financial support and Dr. A Kondilis for critical reading of the manuscript.

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