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Midinfrared scattering and absorption in Ge powder close to the Anderson localization transition

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Using a free electron laser we have performed midinfrared transmission and reflection measurements in strongly scattering Ge powder. We have studied the wavelength dependence of the scattering properties and the optical absorption. Results indicate that our samples are close ($kl_s \simeq 3$) to the Anderson localization transition but still above it. This is in contrast to the recently reported strong localization of light ($kl_s < 1$) in a similar powder of lower refractive index particles (GaAs).

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The similarities in the propagation of electron waves and classical waves have led to an increasing interest in the research of the transport of light in disordered scattering systems [1]. The final goal of many of these studies has been the observation of Anderson localization of light. Anderson localization refers to a breakdown of wave propagation due to constructive interference [2]. In three-dimensional systems, the transition between extended states and localized states occurs when $kl_s \simeq 1$ [3], where k is the wave vector in the medium and l_s the scattering mean free path. The wave vector is given by $k = 2\pi n_e/\lambda$, where n_e is the effective refractive index of the medium and λ the optical wavelength. To approach the localization transition l_s needs to be reduced, which is accomplished by maximizing the scattering cross section σ_s. For light, σ_s is larger when the ratio between the refractive indexes of the scatterers (n) and the surrounding medium is high and when the size of the scatterers is of the order of the wavelength. Although it is a complicating factor, the presence of optical absorption is not necessarily a disadvantage. The role of absorption in the localization process is still under debate [4], and it is unique to optical systems, since absorption is not present where the number of electrons is conserved.

Difficulties in realizing a random medium where the scattering is efficient enough to induce localization has been the reason why only few works report three-dimensional localization of electromagnetic waves [5,6]. In Ref. [6] strong localization ($kl_s < 1$) of near infrared light was reported in GaAs ($n = 3.5$) powder samples consisting of randomly packed micron-size particles. However, an alternative explanation for these measurements has been proposed in terms of classical light diffusion and optical absorption [7]. Clearly, a thorough characterization of the scattering and absorption is needed to understand the transport of light in strongly scattering media. In this Rapid Communication, we present a systematic study of the wavelength dependence of the scattering properties and the optical absorption in Ge powders close to the Anderson localization transition.

The refractive index of Ge is very high in the near and midinfrared ($n \simeq 4.0$) [8], and the absorption of intrinsic Ge very low in the wavelength range 2–15 μm. Therefore, Ge is a very good candidate to prepare a material where infrared light can be localized. We performed three distinct experiments on the Ge powders: midinfrared coherent transmission and diffuse total transmission and reflection measurements in the wavelength range 4.5–8 μm. From the coherent transmission we obtain l_s, while the diffuse transmission and reflection measurements give us the transport mean free path, l, or the average length over which the direction of propagation of light is randomized due to scattering. Our samples are very close to the transition where localization effects are expected to occur. As the transition is approached l is expected to be reduced due to interference, becoming shorter than l_s. However, we did not observe evidence of strong localization in our samples in spite of the very high refractive index of Ge.

The propagation of light, in the weak scattering limit $kl_s \gg 1$, in optically thick disordered scattering media is well described by the diffusion equation. The diffusion equation neglects the interference of waves propagating along different paths since on average this interference cancels out. The light diffuses in the medium with a Boltzman diffusion constant given by $D_B = v_e l_B/3$, where v_e is the energy velocity and l_B is the Boltzman mean free path or the transport mean free path in the absence of interference. In the weak scattering limit, the Boltzman and the scattering mean free paths are related by the expression $l_B = l_s/(1 - \langle \cos\vartheta \rangle)$, where $\langle \cos\vartheta \rangle$ is the average of the cosine of the scattering angle. Notice that l_B is equal to or larger than l_s, which is not surprising since at least one scattering event is required to randomize the direction of propagation of the light. If the absorption length, L_a, is much larger than the sample thickness, L, the total diffuse transmission scales with l/L, similarly to Ohm's law for the electronic conductance. If $L_a < L$ the diffuse transmission decreases exponentially with the sample thickness.

As l_s approaches the critical value at which the Anderson localization transition takes place, the diffusion constant is

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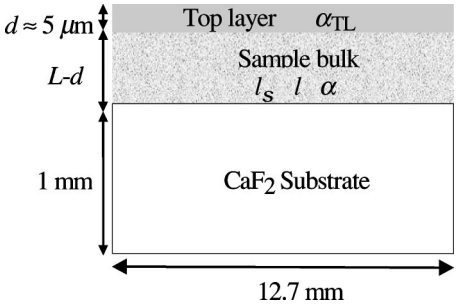

FIG. 1. Schematic representation of a Ge powder sample.

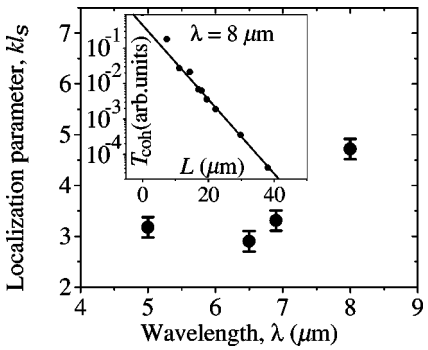

FIG. 2. Localization parameter, kl_s, in Ge powder samples as a function of the wavelength, λ. Inset: Measurements of the transmission of the coherent beam ($\lambda = 8\ \mu$m) as a function of the sample thickness, L. The line is an exponential fit to the measurements from which we obtain $l_s = 3.8 \pm 0.2\ \mu$m.

renormalized by wave interference. According to the scaling theory of localization [1,9], the renormalization of the diffusion constant may be expressed as a reduction of the transport mean free path and, for isotropic scatterers, it is given by

$$\frac{D}{D_B} = \frac{l}{l_s} = l_s\left(\frac{1}{\xi_0} + \frac{1}{L_a} + \frac{1}{L}\right), \tag{1}$$

where ξ_0 is the coherence length. Close to the transition l becomes smaller than l_s. At the transition the coherence length diverges and the transport mean free path vanishes in an infinite and nonabsorbing medium. Therefore, the measurements of both mean free paths give insightful information about the Anderson localization transition, since an l shorter than l_s can be attributed to localization effects. Equation (1) is valid for a cube-geometry sample where the cutoff lengths for ξ_0 due to absorption and the finite size of the sample are included at the same level. Most of the experiments in random media of scatterers are done in slab-geometry samples in which the x and y dimensions are much larger that the z dimension. For such samples the contribution of absorption in Eq. (1) is expected to be more important than the finite thickness of the sample, since light paths longer than L_a are removed due to absorption, while paths much longer than L are still possible along the x-y planes.

Our samples are layers of varying thickness of close-packed Ge micron-size particles on CaF_2 substrates. A detailed description of the sample preparation will be published elsewhere [10]. To estimate the particle size and to check the homogeneity of the samples we took scanning electron microscopy (SEM) images. In Fig. 1 a sample is schematically represented. The samples are formed by a thin top layer of small particles with a linear dimension of $0.19 \pm 0.13\ \mu$m. The thickness of this layer, d, is $5 \pm 1\ \mu$m and it is constant for all the samples. By means of energy dispersion x-ray spectroscopy we have verified that the top layer is mainly formed by Ge particles together with some impurities introduced during the sample preparation. Due to the small size of the particles in this layer the scattering is negligible. Therefore, in the top layer only absorption takes place, characterized by the absorption coefficient α_{TL}. The bulk of the sample, with a thickness L-d (see Fig. 1), is formed by bigger Ge particles with a dimension of $0.98 \pm 0.68\ \mu$m. High resolution SEM images at different places of the bulk showed that the size distribution of the Ge particles was the same at different locations within the same sample, and in different samples. In the sample bulk scattering and absorption take place. The scattering is characterized by the scattering (l_s) and transport (l) mean free paths, and the absorption by the absorption coefficient α. Due to the irregular shape of the Ge particles in the sample bulk, their size and their random orientation, we may consider the scattering to be nearly isotropic.

The radiation source for our optical experiments was a free-electron laser (FEL) (FELIX, Rijnhuizen, The Netherlands) [11]. The FEL is continuously tunable in the midinfrared and delivers a train of picosecond pulses of $\simeq 1\ \mu$J energy. Each train, called macropulse, contains about 100 micropulses. In general the signal from 25 macropulses is averaged at each wavelength. In addition, a reference detector before the sample was used to correct for intensity fluctuations of the laser.

To obtain l_s we measured the decay in intensity of the coherent beam transmitted by the sample as a function of sample thickness. The transmitted fraction of the coherent beam is given by

$$T_{\mathrm{coh}} \propto e^{-(L-d)/l_{ext}}, \tag{2}$$

where l_{ext} is the extinction mean free path [1]. For weak absorption in the sample bulk ($l_s \ll \alpha$) l_{ext} equals l_s. The coherent transmission was detected with a mercury cadmium telluride (MCT) detector placed at a distance of 120 cm from the sample and in the direction of the incoming FEL beam. We measured the transmission of the coherent beam at four wavelengths, 5, 6.5, 6.9, and 8 μm. In the inset of Fig. 2 we plot the measurements at $\lambda = 8\ \mu$m as a function of the sample thickness. From a fit to Eq. (2) (shown by the solid line), a value of $l_s = 3.8 \pm 0.2\ \mu$m is obtained for this λ. To obtain kl_s we need the effective refractive index of the samples, n_e. A good estimate of n_e is given by the Maxwell-Garnet effective refractive index [12]. Considering a Ge volume fraction of 40% [6,13] we find $n_e \approx 1.6$ in the measured λ range. Figure 2 shows the wavelength dependence of kl_s in the Ge powder samples. The high polydispersity in the particle size is responsible of the nearly constant value of $kl_s \simeq 3$ at $\lambda = 5$, 6.5, and 7 μm [13]. However, at higher wavelengths kl_s becomes larger, which is expected since the scattering cross section is reduced. The low kl_s is an evidence of the very strong scattering in our samples.

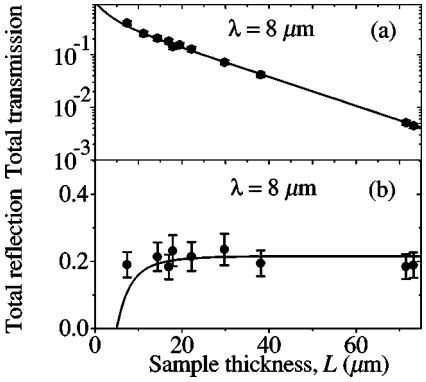

FIG. 3. Total transmission (a) and reflection (b) measurements in Ge powder samples at $\lambda = 8\ \mu$m as a function of the samples thickness, L. The solid lines represent fits to the measurements using the diffusion theory.

To look for localization effects it is necessary to measure a transport parameter such as l. The transport mean free path can be obtained from the diffuse total transmission and reflection, defined as the transmitted and reflected fluxes normalized by the incident flux. The total transmission was measured by collecting the transmitted light with an infragold-coated integrating sphere (IS). For the total reflection measurements the FEL beam was sent into the IS through a small input port. The sample was at the opposite side of the IS, which collected only the reflected light. The total transmission and reflection measurements at $\lambda = 8\ \mu$m as a function of the sample thickness are plotted in Figs. 3(a) and (b) respectively.

For our samples the total transmission, T, is given by

$$T = e^{-\alpha_{\mathrm{TL}} d}(T_d + e^{-(L-d)/l_s}). \tag{3}$$

The factor $e^{-\alpha_{\mathrm{TL}} d}$ represents the attenuation of the incoming beam due to absorption in the top layer. The first term inside the bracket, T_d, is the diffuse total transmission, while the second term is the transmission of the coherent beam. The total reflection, R, is more sensitive than the total transmission to the absorption in the sample top layer because the reflected light crosses this layer two times. The total reflection is given by

$$R = e^{-\alpha_{\mathrm{TL}} d} R_d \int_0^1 e^{-\alpha_{\mathrm{TL}} d/\mu} P(\mu)\, d\mu, \tag{4}$$

where R_d is the diffuse reflection of the sample bulk, $\mu = \cos\theta$ and θ is the angle with respect to sample surface normal at which the diffuse reflected light exits the sample bulk. The diffuse reflected light is angular distributed according to $P(\mu) \propto \mu[\frac{2}{3} + (\mu/1+\mu)]$ [14], where we assume that the bulk of the sample and the top layer have the same index of refraction; this is justified since we know that the top layer is mainly formed by Ge and from the SEM images that the packing of the particles is similar in both regions. The integral in Eq. (4) represents the attenuation of the diffuse reflected light due to absorption in the top layer. The diffuse total transmission, T_d, and reflection, R_d, can be calculated in the weak scattering limit and close to the localization transition (but still above it) by solving the diffusion equation.

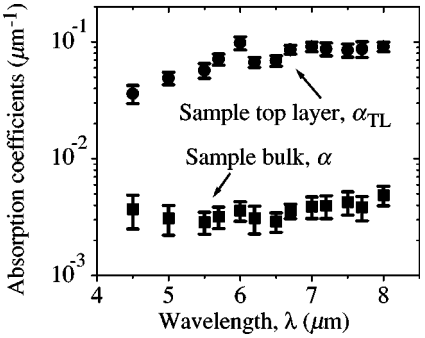

FIG. 4. Absorption coefficients in the Ge powder samples top layer, α_{TL}, and in the bulk, α, as a function of the wavelength, λ.

The boundary conditions of the diffusion equation are determined considering that the diffuse fluxes going into the sample are due to a finite reflectivity at the boundaries [15]. T_d and R_d depend on the reflectivity at the boundaries by means of the so-called extrapolation factors τ_1 and τ_2 [15], where index 1 refers to the boundary where the incident beam enters the sample and index 2 to the opposite boundary. Knowing the effective refractive index of the sample and refractive index of the substrate, τ_2 can be easily calculated [16]. The extrapolation factor τ_1 will depend on α_{TL} and it can be obtained from the total transmission measurements [10].

The solid lines in Fig. 3(a) and 3(b) are fits to the total transmission and reflection measurements ($\lambda = 8\ \mu$m) using Eqs. (3) and (4). From the fits we obtain at this particular wavelength $l = 3.5 \pm 0.5\ \mu$m, $L_a = 15.5 \pm 1\ \mu$m, $\alpha_{\mathrm{TL}} = 0.091 \pm 0.008\ \mu\mathrm{m}^{-1}$, and $\tau_1 = 1.6 \pm 0.8$. The absorption coefficient in the sample bulk is given by $\alpha = l/(3L_a^2)$. The absorption coefficients α_{TL} and α are plotted in Fig. 4 as a function of the wavelength. Clearly, significant absorption has been introduced during the sample preparation. The absorption is more intense in the top layer where the size of the particles is smaller and where we have measured the presence of impurities.

In Fig. 5 we display l/l_s as circles. The values of l/l_s are smaller than 1, which can be only understood by considering the renormalization of l due to the proximity of the localization transition. Far from the transition l/l_s gives the average

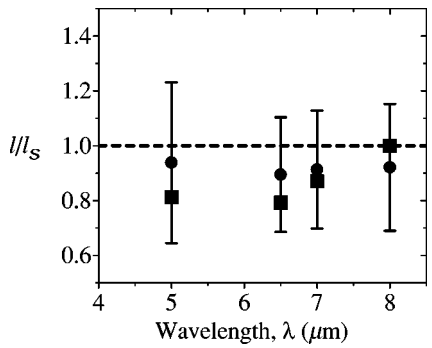

FIG. 5. Circles: transport mean free path, l, of light of wavelength λ in the Ge powder samples, divided by the scattering mean free path, l_s. Squares: Expected renormalization of l due to interference assuming that the localization transition is at $l_s = \lambda/2\pi n_{\mathrm{e}}$ and $n_{\mathrm{e}} = 1.6$.

number of scattering events required to randomize the direction of propagation of the light. Therefore, in this limit l/l_s is always equal to (isotropic scatterers) or greater than (anysotropic) 1. For comparison we also plot in Fig. 5 (squares) the expected renormalization of l according to the scaling theory of localization, Eq. (1), assuming that the transition occurs at $kl_s=1$ and considering only the absorption as cut-off of ξ_0. Good agreement between experiments and theory is found. However, the large error in the determination of l, related to the complicated structure of the samples and to the estimation of the boundary reflectivities, makes it impossible to unambiguously exclude localization effects in the Ge samples. From the results shown in Figs. 2 and 5, we may conclude that our Ge samples are very close to the localization transition but still above it. This is a surprising result since strong localization ($kl_s<1$) of light has been reported in similar samples of GaAs powder [6]. GaAs has a lower refractive index than Ge and therefore the scattering efficiency in GaAs powder is expected to be lower than in Ge samples. At this moment it is not clear why strong localization is apparent in GaAs powder while it is absent in Ge powder. More experimental and theoretical work is needed to confirm the results of Ref. [6]. A way to further approach the localization transition in the Ge samples could be to reduce the polydispersity in the particle size, as discussed in Ref. [13].

In conclusion, we have studied the wavelength dependence of the scattering properties of Ge powder, using the midinfrared light from a free-electron laser. By measuring the coherent transmission of the laser beam, we obtain the scattering mean free path, l_s. Our samples are close to the localization transition but still above it. This is an unexpected result since Anderson localization of light has been reported in similar samples of lower refractive index material, GaAs [6]. We have also measured the total transmission and reflection of the Ge powder samples obtaining the transport mean free path, l, and the optical absorption in the samples. During the sample preparation we have introduced significant absorption. Our measurements suggest a renormalization of l due to the proximity to the localization transition.

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