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Infrared probe of metal-insulator transition in $Si_{1-x}Gd_x$ and $Si_{1-x}Y_x$ amorphous alloys in magnetic field

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Abstract. – Carrier dynamics in amorphous a-$Si_{1-x}RE_x$ (RE = Gd, Y) films has been studied in the doping regime close to the metal-insulator transition by means of infrared spectroscopy. Optical constants throughout the entire intra-gap region ($\hbar\omega < 1\,\mathrm{eV}$) have been found to be anomalously sensitive to changes of temperature and/or magnetic field. The observed behavior is consistent with the model of hopping transport where the interaction of carriers with both the lattice and large core spin of Gd ions is taken into account.

Magnetic dopants may radically alter the properties of prototype semiconductors, such as Si or GaAs, and may lead to extraordinary effects even in these fairly simple systems [1–3]. For example, III-V materials doped with Mn develop ferromagnetism with the Curie temperature exceeding 110 K [1] and amorphous Si alloyed with a high concentration of Gd shows gigantic negative magnetoresistance [3]. Present understanding of the role of magnetic impurities in carriers dynamics, especially in the regime where a material is close to the boundary of the metal-insulator transition (MIT), is rather poor, in part because the experimental results are very scarce. This motivated us to perform studies of the electromagnetic response of a-$Si_{1-x}Gd_x$ in the vicinity of the MIT using the techniques of infrared (IR) optics and magneto-optics, which are ideally suited for the task.

We have measured the IR spectra of heavily doped a-$Si_{1-x}Gd_x$ and its non-magnetic counterpart a-$Si_{1-x}Y_x$ close to the boundary of the metal-insulator transition. The amorphous Si matrix allows one to increase the concentration of magnetic Gd ions beyond the solubility limit in crystalline silicon. At $x \simeq 0.11$–0.13 these materials are insulating at $T \to 0$ in zero magnetic field but become conducting in the field $H \simeq 5\,\mathrm{T}$ [3]. The effect is reminiscent of the behavior of diluted magnetic semiconductors [4]. However, in the latter case negative

magnetoresistance (MR) is usually observed close to a temperature of magnetic ordering [4]. On the contrary, a-$Si_{1-x}Gd_x$ alloys show large negative MR at all temperatures below $\simeq 80\,\mathrm{K}$, and the magnitude of the MR is not affected by spin-glass–like freezing at $T < T_{\mathrm{sg}} = 5$–$10\,\mathrm{K}$. It is worth mentioning that the standard theory of electron hopping predicts *positive* MR in amorphous semiconductors [5], whereas a-$Si_{1-x}Gd_x$ exhibits huge *negative* MR. We have found that one particular signature of the MIT in these systems is associated with the massive transfer of the spectral weight over an energy interval exceeding 1 eV. This scale is larger than the typical magnitudes of the Coulomb gap or the correlation gaps forming at the chemical potential in the density of states in the insulating samples [5,6]. At the same time the interaction of carriers with the lattice is known to be particularly strong in amorphous solids [7,8] and, together with exchange coupling of carriers to Gd local spins, it allows us to account for the optical properties of a-$Si_{1-x}Gd_x$.

The transmission coefficient $T(\omega)$ over the frequency interval 20–10000 cm^{-1} has been measured for $\simeq 0.5\,\mu$m a-$Si_{1-x}Gd_x$ films prepared by electron beam co-evaporation [3] on a high-resistance ($\sim 10\,\mathrm{k}\Omega\,\mathrm{cm}$) Si substrate. Zero-field measurements have been carried out using a Michelson interferometer equipped with He-cooled bolometers for the far-IR studies and with the HgCdTe detector for spectroscopy in the mid-IR range. These experiments were supplemented with the studies of the changes of the 10 K transmission in magnetic field up to 15 T. For these latter studies we employed a bolometer as a detector with different combinations of cut-off filters allowing us to collect data for ω as high as $4500\,\mathrm{cm}^{-1}$. The absolute accuracy of our measurements was about 1%, whereas the changes of transmission as a function of temperature or field are characterized with the relative accuracy better than 0.2%. Here we report the data for the "critical" sample with 12 at.% Gd, in which the dc conductivity σ_{dc} vanishes at $T \to 0$, and for a "metallic" sample with 15 at.% Gd in which $\sigma_{\mathrm{dc}}(T \to 0) \simeq 80\,\Omega^{-1}\mathrm{cm}^{-1}$ increases up to $300\,\Omega^{-1}\mathrm{cm}^{-1}$ at $T = 300\,\mathrm{K}$. In order to isolate the effects associated with magnetic interactions, we compare the Gd-doped samples (core f-spin $S = 7/2$) with films containing a similar amount of non-magnetic Y. Complex conductivity $\sigma_1(\omega) + i\sigma_2(\omega)$ was determined from the Kramers-Kronig (KK) analysis of $T(\omega)$ following the standard procedure [9].

The dominant feature of the $\sigma_1(\omega)$ spectra of all films is an absorption edge around $10000\,\mathrm{cm}^{-1}$, fig. 1. This feature can be attributed to the gap in the density of states of amorphous Si. We have also found significant absorption throughout the intragap region that is dramatically affected by changes in temperature and magnetic field. To quantify the strength of the intragap absorption, we define the effective spectral weight [10]

$$N_{\mathrm{eff}}(\omega) = \int_0^{\omega} \mathrm{d}\omega' \sigma_1(\omega'). \tag{1}$$

The magnitude of N_{eff} is proportional to n/m^*, where n is the density of carriers participating in the conductivity at energies below ω and m^* is their mass. The integration up to $\omega = 5000\,\mathrm{cm}^{-1}$ [11] gives the following estimates of the carrier density, under the assumption that $m^* = m_{\mathrm{e}}$: $n = 3.9 \times 10^{20}\,\mathrm{cm}^{-3}$ for the critical sample and $n = 5.4 \times 10^{20}\,\mathrm{cm}^{-3}$ for metallic samples.

All films reveal a broad resonance at $\omega \simeq 700$–$1000\,\mathrm{cm}^{-1}$; the resonance becomes more pronounced with increasing concentration of dopants. This feature is characteristic of hopping conductivity in disordered semiconductors, as will be discussed below. The absolute value of the $\sigma_1(\omega)$ in the far-IR region is strongly suppressed at low T [12]. Because of this suppression of the conductivity, part of the far-IR spectral weight appears to be lost. Integration of the $\sigma_1(\omega)$ spectra shows that the missing weight is not recovered at $\omega < 1\,\mathrm{eV}$. According to

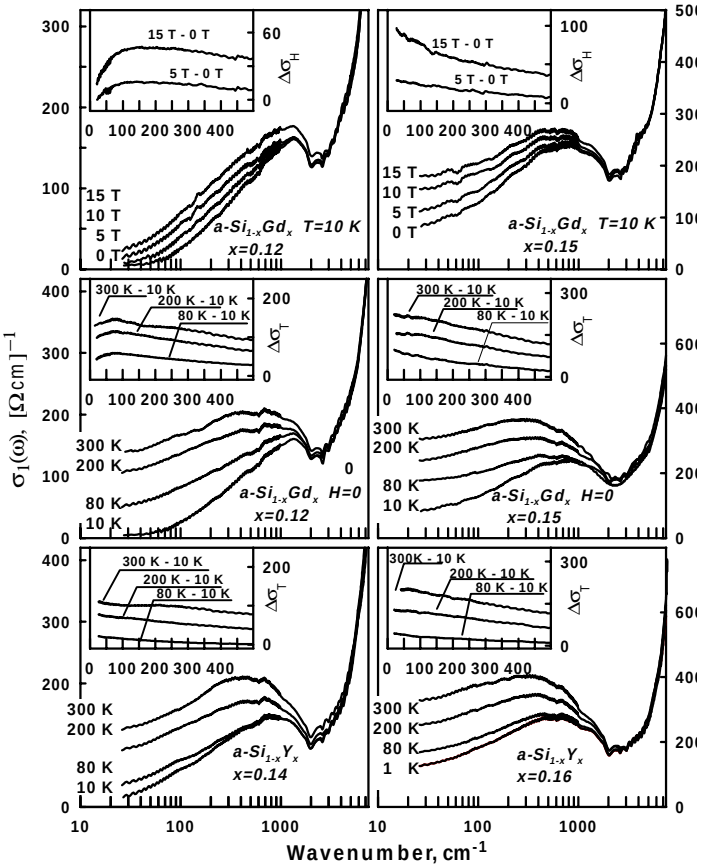

Fig. 1 – The optical conductivity of a-$Si_{1-x}RE_x$ films at different temperatures (middle and bottom panels) and magnetic fields (top panels). Insets show the differential conductivity $\Delta\sigma_T(\omega)$ and $\Delta\sigma_H(\omega)$ defined in the text. In all samples the intragap conductivity is strongly reduced at low T with the spectral weight being transferred to energies exceeding the band gap of amorphous Si. In a-$Si_{1-x}Gd_x$ films a part of the spectral weight, which is lost at $T < 80\,\mathrm{K}$, is recovered in high magnetic field (top panels).

the f-sum rule, $N_{\mathrm{eff}}(\infty) = n/m_{\mathrm{e}} = \mathrm{const}$, so the total area under the $\sigma_1(\omega)$ spectrum must remain constant. We conclude, therefore, that the spectral weight disappearing from the intragap region is transferred to frequencies above the gap value. This result is anomalous since changes of temperature by 200–300 K have a conspicuous impact on the spectra of $\sigma_1(\omega)$ over the energy interval exceeding 1 eV (12000 K). We are unaware of an accurate analysis of the oscillator strength throughout the intragap region in other amorphous semiconductors. Unusual transfer of the spectral weight is known to occur in systems with strong correlations between the electrons [4,13]. We emphasize that at $T > 80\,\mathrm{K}$ both Y- and Gd-doped samples show a similar behavior suggesting that these effects are common in both magnetic and nonmagnetic alloys.

The decline in the conductivity of a-$Si_{1-x}Gd_x$ samples continues at low temperatures $\lesssim$ 70–80 K; the conductivity of a-$Si_{1-x}Y_x$ films is only weakly temperature-dependent. This additional reduction of the conductivity drives the critical Gd-doped sample towards the

insulating state, whereas the a-$Si_{1-x}Y_x$ film with similar doping remains "metallic": $\sigma_{dc}(T \to 0) \approx 90\,\Omega^{-1}cm^{-1}$. The spectral weight, lost from the far-IR region at $T < 80\,K$ in a-$Si_{1-x}Gd_x$, is also transferred to energies beyond 1 eV.

Changes in the conductivity of a-$Si_{1-x}Gd_x$ films induced by magnetic field are even more anomalous. In applied field both metallic and critical films recover most of the spectral weight, which was lost from the conductivity at $T < 80\,K$. Field-induced enhancement of N_{eff} is directly related to the large negative MR found in Gd-based films. Remarkably, the magnetoconductivity $\Delta\sigma_H = \sigma_1(\omega, H) - \sigma_1(\omega, H = 0)$ of both "critical" and "metallic" films remains positive up to at least 0.5 eV. This frequency region exceeds the energy scale associated with the magnetic field by more than 2 orders of magnitude. IR magnetoconductivity of Y-based films is negligibly small.

Insets in fig. 1 show the $\Delta\sigma_H$ and $\Delta\sigma_T = \sigma_1(\omega, T) - \sigma_1(\omega, 10\,K)$ spectra indicating important distinctions in the response of magnetic and non-magnetic films. The differential spectra show a monotonic ("Drude"-like) dependence in Y-based samples and in the metallic Gd-based samples. In contrast, both $\Delta\sigma_T$ and $\Delta\sigma_H$ for the critical a-$Si_{1-x}Gd_x$ film display a peak centered at around 10 meV. This latter behavior is characteristic of disordered semiconductors, where carriers are localized, with the peak position corresponding to the magnitude of the mobility gap [14]. The 10 meV feature may account for the enhancement of localization in magnetic Gd-based films. Interestingly, the energy associated with the peak agrees quantitatively with the temperature at which the localization effects become prominent in the transport measurements. Indeed, the dc conductivities of a-$Si_{1-x}Gd_x$ and a-$Si_{1-x}Y_x$ films for $x \simeq 0.11$–0.13 are identical at $T > 80\,K$. However, below this temperature $\sigma_{dc}(T)$ in a-$Si_{1-x}Gd_x$ is suppressed faster than that of Y-based film and eventually vanishes at $T \to 0$. On the contrary, the Y-based samples remain "metallic" [3]. It is reasonable to ascribe the difference between a-$Si_{1-x}Gd_x$ and a-$Si_{1-x}Y_x$ films to magnetic exchange interactions, which are present only in the Gd-based system. We can estimate the exchange coupling between the core f-spin and the carrier spin as $JS \sim 10$ meV [15], which is in good agreement with localization features seen both in transport and IR experiments. We do not observe the corresponding peak in the spectra for *metallic* Gd-based films. The localization trends are much less pronounced in this sample, and one can speculate that the mobility gap is shifted there to lower energies, so that the signature of this effect in $\sigma_1(\omega)$ occurs below the experimental low-ω cutoff.

We emphasize that the energy scale involved in the transfer of the spectral weight with decreasing T and partial recovery of N_{eff} with increasing field H extends beyond 1 eV. Therefore, this scale dramatically exceeds the mobility gap in the studied films as well as the Zeeman energy for a-$Si_{1-x}Gd_x$ by at least two orders of magnitude. Looking for possible origins of the effects, extending over such unexpectedly large energy scale, one can recall that the shift of the electronic levels to lower energies due to interaction with the lattice (the so-called polaron shift E_p) can readily reach 1 eV in many semiconductors [16]. A strong electron-lattice interaction is favored by the "softness" of an amorphous matrix and leads to the formation of lattice polarons and local singlet pairs (Anderson bipolarons) [7,8]. To illustrate a possible effect of lattice (bi)polarons, we consider the following model. We assume that the energies of localized carriers at site $\boldsymbol{i}$ and spin $s = \uparrow (\downarrow)$ are split by the external field H and the exchange interaction with the core f-spins S ($S = 7/2$ for Gd and 0 for Y)

$$\epsilon_{\boldsymbol{i}\uparrow(\downarrow)} = \epsilon^0_{\boldsymbol{i}} - (+)\frac{1}{2}JS\eta - (+)\mu_B H, \tag{2}$$

where $\eta = \langle S_z \rangle / S$ is the relative f-spin polarization, $JS \simeq 10$ meV as discussed above, μ_B is the Bohr magneton, the impurity levels $\epsilon^0_{\boldsymbol{i}}$ are distributed with the width Γ (~ 0.2 eV [17]).

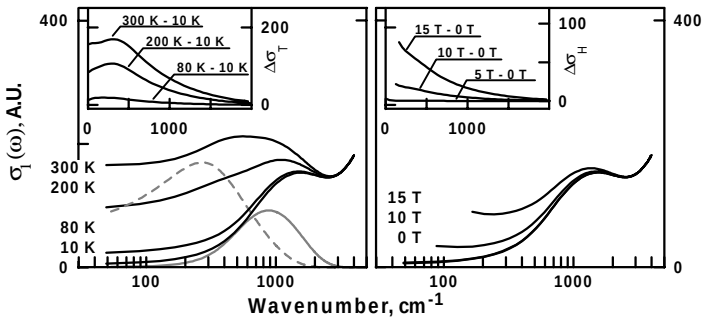

Fig. 2 – The calculated optical conductivity $\sigma(\omega; T, H)$ of a-$Si_{1-x}Gd_x$ for $x = 0.12$ at different temperatures and magnetic fields. Gray lines in the left panel show separate contributions to the conductivity associated with the polarons (dashed line) and spin pairs (solid line) at $T = 300\,\mathrm{K}$. The polaron absorption peak is at $\omega_\mathrm{p} = 200\,\mathrm{cm}^{-1}$. The insets show $\Delta\sigma_T(\omega)$ and $\Delta\sigma_H(\omega)$. The model spectra reproduce the key features of the experimental $\sigma(\omega)$ presented in fig. 1, as well as the redistribution of the spectral weight induced by temperature (left panel) and magnetic field (right panel).

This splitting results in the polarization of the carriers (lattice polarons) $\mu = n_{\mathrm{p}\uparrow} - n_{\mathrm{p}\downarrow}$, where $n_{\mathrm{p}s}$ is the density of polarons with spin $s = \uparrow (\downarrow)$. Within this picture, the pairs constitute the ground state, with the individual polarons existing as thermal excitations [7,8]. The electron-phonon coupling in amorphous semiconductors is known to be sufficiently strong to bind carriers into singlet spin pairs with the binding energy $\Delta \sim 30\,\mathrm{meV}$, given by the difference between electron-phonon pairing and Coulomb on-site repulsion [7]. An estimate for Δ in the studied systems can be found from the magnitude of the "hard" gap seen in tunneling data [6].

Both single-lattice polarons and bound pairs produce the Gaussian-like absorption resonance $\sigma_1(\omega)$ [16,18,19]. The conductivity due to excitation of single polarons is peaked at ω_p, which is proportional to E_p [19], and has the form $\sigma_\mathrm{p}(\omega) \propto [n_{\mathrm{p}s}/\omega\gamma] \exp[-(\omega-\omega_\mathrm{p})^2/\gamma^2]$, where $\gamma(\approx 30\,\mathrm{meV})$ is the width of the absorption resonance. Analogously, the conductivity of the bound pairs is described as $\sigma_\mathrm{bp}(\omega) \propto [n_\mathrm{bp}/\omega\gamma] \exp[-(\omega-\omega_\mathrm{bp})^2/\gamma^2]$, where n_bp is their density and $\omega_\mathrm{bp} = \omega_\mathrm{p} + \Delta$, with Δ being the binding energy of the pairs [16,18,19]. The scatter of the energies of the impurity states, Γ, results in the observed asymmetric shape of the absorption line [18]. The hopping conductivity is the sum of both contributions: $\sigma_\mathrm{hopping} = \sigma_\mathrm{p} + \sigma_\mathrm{bp}$. In order to calculate the spectra presented in fig. 2, we add an interband contribution σ_inter to σ_hopping. The standard form for σ_inter in amorphous semiconductors is $\sigma_\mathrm{inter} \propto (\omega - \omega_0)^2/\omega$, with the threshold energy $\omega_0 \approx 0$ [20]. The calculated conductivity $\sigma_\mathrm{hopping}(\omega) + \sigma_\mathrm{inter}(\omega)$ at different T (fig. 2, left panel) is in fair agreement with the experimental data. In particular, the model accounts for the massive transfer of the spectral weight observed in our films at low temperatures. This is due to the increase of the density of single lattice polarons $n_\mathrm{p} \propto \exp[-(\Delta - JS\eta - 2\mu_\mathrm{B}H)/2k_\mathrm{B}T]$ with temperature as the result of thermal dissociation of the local spin pairs. An important feature of this model is that a fraction of the spectral weight associated with the higher-order hopping processes appears at energies comparable to the polaron shift. Although the detailed account for these processes is beyond the saddle-point approximation used to estimate σ_hopping [18,19], our model shows that at higher T their contribution is reduced, leading to an enhancement of the low-energy spectral weight.

Another feature of the polaronic scenario is that it results in the negative optical magnetoresistivity observed in a-$Si_{1-x}Gd_x$, provided that the exchange coupling of carriers with a large core f-spin S of Gd is taken into account. According to eq. (2), lattice polarons

are polarized by the external field H *and* by a much stronger molecular field $JS\eta/\mu_{\rm B}$. Their polarization μ produces the exchange field $J\mu/(2g_{\rm Gd}\mu_{\rm B})$ which in return polarizes the f-spins S_z. The polarizations η and μ are determined self-consistently, analogously to ref. [16]. The exchange interaction strongly enhances the polarization of carriers μ in the external field. Since the spin of lattice polarons interacts with the exchange and external fields, the polaron conductivity of Gd-doped films $\sigma_{\rm p}(\omega)$ acquires a field-dependent factor, $\sigma_{\rm p}(\omega, H) = \sigma_{\rm p}(\omega, 0)\cosh[(\frac{1}{2}JS\eta + \mu_{\rm B}H)/k_{\rm B}T]$. This term increases rapidly with the applied field, as does the density of polarons. Note that "magnetic" polarons do *not* form in our systems [21]. The net results are displayed in fig. 2. One finds a strong enhancement of the $\sigma_{\rm p}(\omega)$ contribution to the total hopping conductivity and very large negative MR due to the transfer of the spectral weight to lower frequencies. While some models were suggested for the spectral weight transfer with the onset of ferromagnetism [4, 22], the novelty of the proposed polaronic scenario is that the effect occurs in the *paramagnetic* state. Thus, the polaronic model introduced above not only reproduces the principal features of the conductivity spectra but also accounts for the non-trivial behavior of the spectral weight at finite temperature and/or field. This agreement highlights the prominent role of the electron-lattice interaction in the correlated behavior of doped amorphous silicon, which leads to the formation of the lattice (bi)polarons [21].

An important open issue concerns a possible coexistence of both extended and localized states on the metallic side of the transition [23]. Indeed, the frequency dependence of the $\sigma_1(\omega, T = 0)$ spectra of the "metallic" samples is similar to what is observed at finite T or H in the optical response of the critical samples. As shown above, these latter regimes are consistent with the coexistence of both extended and localized states at the chemical potential, indicating that a similar co-existence may occur at $T = H = 0$ on the "metallic" side of the transition. We also note that the proposed polaronic scenario requires the presence of nonbound carriers at $T \to 0$ even in the critical films in order to explain the negative MR of a-$Si_{1-x}Gd_x$ at very low temperatures [3]. This requirement stems from the fact that the magnetoresistance is related to the existence of unpaired polarons, since they only are subject to the exchange interaction with the core spins on the Gd sites. We conclude by noting that the properties of a-$Si_{1-x}Gd_x$ alloys studied in the present work with the use of IR spectroscopy are likely to be generic to other classes of amorphous semiconductors with magnetic impurities.

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[11] The choice of $\omega_{\rm c} = 5000\,{\rm cm}^{-1}$ is suggested by the frequency dependence of the $\sigma_1(\omega)$ spectra shown in fig. 1. At $\omega > 5000\,{\rm cm}^{-1}$ the conductivity is clearly dominated by transitions across the band gap of Si with significantly broadened band tails in the present amorphous samples.
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