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On the relaxation rate distribution of the photoionized DX centers in indium doped $Cd_{1-x}Mn_xTe$

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Abstract
It was recently shown that the kinetics of persistent photoconductivity (PPC) build-up in indium doped $Cd_{1-x}Mn_xTe$ are non-exponential and can be described solely by the stretched-exponential function. The non-exponentiality is attributed to the indium related DX centers present in the materials. In order to explain this observation, low temperature photoconductivity build-up was studied for $Cd_{1-x}Mn_xTe$:In of two different manganese contents. It was found that this type of response has its origin in the heavy-tailed distribution of the DX centers. The distribution was analyzed in terms of photon flux. Increasing photon flux leads to the more dispersive behavior. It was also confirmed that the heavy-tailed distribution is due to the different local configuration of atoms surrounding DX centers in the alloy.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Investigations of the origin of non-exponential relaxation processes in a wide class of various materials have led to the conclusion that the empirically observed relaxation patterns reflect some universal behavior. It has been established that independently of the considered medium the relaxation response obtained by different experimental techniques can be well characterized by a small class of fitting functions exhibiting asymptotic properties—the so-called power-laws [1–3]. The power-laws are observed in both the time and frequency domain of the measurement and can be easily noted in a double-logarithmic scale since in this scale any power dependence is linear.

Recent development of the stochastic approach to relaxation allowed us to clarify the mechanisms responsible for the experimentally observed short-time power-laws. Within this approach not only the distribution of the relaxation rates can be calculated but also the explicit form of the relaxation function may be derived [4–6]. The clue of the stochastic model is that it allows us to describe the statistical properties of the relaxing entities, i.e. (i) the number of objects contributing to the relaxation response, (ii) the distribution of individual relaxation contributions and (iii) the distribution of the effective relaxation rate [7]. Moreover this scheme brings information concerning interactions among the relaxing entities, i.e. the possibility of their clusterization and interaction among the clusters. An appropriate combination of cluster sizes and distributions of relaxation rates leads to the well-known relaxation responses such as the stretched-exponential Kohlrausch–Williams–Watts (KWW) (in the time-domain) or Havriliak–Negami (in the frequency-domain) functions [8].

The KWW relaxation function given by equation:

$$\Phi_{\mathrm{KWW}}(t) = \mathrm{e}^{-\left(\frac{t}{\tau}\right)^{\alpha}}, \tag{1}$$

where $0 < \alpha < 1$ is the stretching exponent and τ is a time constant of the transient, is the most frequently used to fit the time-domain relaxation data. This function leads to the response function, defined as the minus of the growth rate of the relaxation function $f(t) = -\frac{\mathrm{d}\Phi_{\mathrm{KWW}}(t)}{\mathrm{d}t}$, decaying at short times as $t^{\alpha-1}$, hence the short-time power-law.

The stretched-exponential function has been widely applied to describe the non-exponential kinetics of photoconductivity decay in materials possessing metastable defects

called DX centers [9–11]. The experimental evidence for the presence of DX centers in the material is a persistent photoconductivity effect (PPC) observed at low temperatures—photoinduced conductivity persists for a long time (hours) after turning off the light. Investigations of the persistent photoconductivity build-up in the indium and gallium doped semiconducting multiternary compound $Cd_{1-x}Mn_xTe$ have shown that photoconductivity decay can be properly described by the KWW function. It was found that also the photoconductivity build-up response transients in these materials are non-exponential and exhibit a short-time power-law property. This law is characteristic for the stretched-exponential relaxation pattern [12–14]. Both the persistent photoeffect and a non-exponential photokinetics in these materials have been attributed to the photoionization of DX centers [15–18].

In order to explain the asymptotic behavior of photoconductivity build-up in gallium doped $Cd_{0.99}Mn_{0.01}Te$ the stochastic approach was applied for the first time [13]. It has been shown that the stretched-exponential form of the measured photoconductivity kinetics results from a heavy-tailed (broad) distribution of the DX centers relaxation rates having its origin in a different local atomic arrangement in this multiternary compound [14]. In the present paper the stochastic model was used to explain a photoconductivity build-up in indium doped $Cd_{1-x}Mn_xTe$. It has been found that this model leads to conclusions similar to those found for gallium doped $Cd_{1-x}Mn_xTe$. In order to shed light on the microscopic behavior of DX centers in $Cd_{1-x}Mn_xTe$ the following problems have been analyzed: (i) the influence of photon flux on the statistical properties of DX centers, (ii) the distributions of the DX centers' relaxation rates, (iii) the correlation among the centers.

2. Stochastic approach to the time domain relaxation processes

Application of the stochastic approach to investigate the relaxation processes allows us to characterize the considered material both micro- and macroscopically and to find a relationship between both points of view [5, 19]. Within this approach a random variable $\beta_i = \frac{1}{\tau_i}$ is assigned to each individual relaxation rate connected with a microscopic relaxation contribution (here—a DX center). Macroscopic properties of the studied medium are represented by a random variable $\tilde{\beta}$, called the effective relaxation rate, equal to the sum of individual rates:

$$\tilde{\beta} = \lim_{N\to\infty} \sum_{i=1}^{N} \frac{\beta_i}{A_N}. \tag{2}$$

In the above formula A_N is a positive scaling constant. It can be shown, using the limit theorems of probability theory [20], that the above limit exists solely if the distribution of the individual relaxation rates β_i belongs to the domain of attraction of the α-stable law. It means that the probability density function of individual rates exhibits the power-law tail for large b:

$$\rho_{\beta_i}(b_i) \sim b_i^{-\alpha-1}, \qquad b_i \to \infty, \quad 0 < \alpha < 1, \tag{3}$$

where $b_i \in [0, \infty)$ denotes values taken by random variable β_i. In probability theory, distributions of non-exponentially decaying tails are called heavy-tailed (long-tailed, power-tailed) distributions [22–25]. The distribution of any non-negative random variable X possesses a heavy-tail if for large values of the support x the tail decays as a fractional power-law $(x/x_0)^{-a}$, where x_0 and $0 < a < 1$ are some positive constants [20].

In the case of the heavy-tailed distribution of β_i the effective relaxation rate $\tilde{\beta}$ is an α-stable random variable $(0 < \alpha < 1)$ $\tilde{\beta}_\alpha$ with an asymmetric probability density function $\rho_{\tilde{\beta}_\alpha}(b)$ which possesses the same power-law tail for large b [6–8, 21]:

$$\rho_{\tilde{\beta}_\alpha}(b) \sim b^{-\alpha-1}, \qquad b \to \infty. \tag{4}$$

In the above relation $b \in [0, \infty)$ represents values taken by the random effective relaxation rate.

Generally, an α-stable density $\rho(b)$ is defined as a Fourier transform of the characteristic function $\varphi(t)$:

$$\rho(b) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \mathrm{e}^{-\mathrm{i}tb} \varphi(t)\, \mathrm{d}t \tag{5}$$

where

$$\log \varphi(t) = \begin{cases} \mathrm{i}\gamma t - c^{\alpha} |t|^{\alpha} \left\{1 - \mathrm{i}\beta \operatorname{sgn}(t) \tan\left(\frac{\pi}{2}\alpha\right)\right\}, & \alpha \neq 1 \\ \mathrm{i}\gamma t - c|t| \left(1 + \mathrm{i}\beta \frac{2}{\pi} \operatorname{sgn}(t) \log|t|\right), & \alpha = 1. \end{cases}$$

The shape of the density $\rho(b)$ is determined by four parameters: the index of stability $0 < \alpha \leqslant 2$, the skewness parameter $-1 \leqslant \beta \leqslant 1$, the location parameter $\gamma \in R$ and the scale parameter $c > 0$ [22, 23]. When α approaches 1, the density tends to the Dirac δ-function. In this case the rate contributions focus around the expected value of their distribution. The relaxing system is deterministic for all relaxing entities' responses to the external disturbance in the same manner. This is the particular case of Debye relaxation.

According to the stochastic definition, a relaxation function is equal to the Laplace transform of the effective relaxation rate probability density function (pdf) $\rho_{\tilde{\beta}_\alpha}(b)$:

$$\Phi(t) = \int_0^{\infty} \mathrm{e}^{-bt} \rho_{\tilde{\beta}_\alpha}(b)\, \mathrm{d}b. \tag{6}$$

The relaxation function defined in equation (6) can be obtained in explicit form as the KWW function $\Phi_{\mathrm{KWW}}(t) = \mathrm{e}^{-(\frac{t}{\tau})^{\alpha}}$ solely if the effective relaxation rate pdf is the asymmetric, right-skewed $(\beta = 1)$ α-stable probability density function with $0 < \alpha < 1$ and the scale parameter $c = [\cos(\frac{\pi\alpha}{2})]^{1/\alpha} \cdot \frac{1}{\tau}$.

To sum up, the α-stable distribution of the effective relaxation rate is a consequence of the heavy-tailed (broad) distribution of individual relaxation rates β_i [21, 26]. In the studied material the individual relaxation rates β_i are assigned to DX centers. The heavy-tailed distributions of the centers mean that some of them respond fast enough to be detected while others do not contribute to the measured photokinetics.

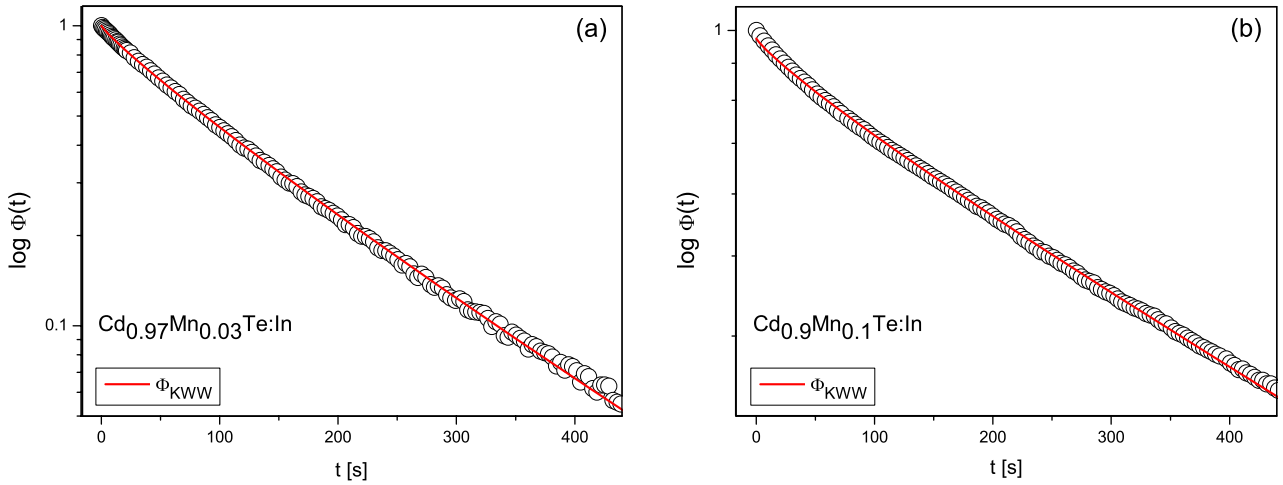

Figure 1. The PPC build-up kinetics normalized according to equation (6). Open circles stand for the experimental data, solid lines for the fitting with the use of the stretched-exponential function Φ_{KWW}.

3. Experiment

Indium doped $Cd_{0.93}Mn_{0.07}Te$ and $Cd_{0.9}Mn_{0.1}Te$ crystals used in this study were grown by the Bridgman method. Prior to the measurements the samples were annealed in cadmium vapor to reduce the cadmium vacancies. Slices of the material were mechanically polished and etched in a 2% Br_2 in methanol solution. Capacitance–voltage measurements performed with a 1 MHz capacitance bridge, yielded a room temperature donor net concentration of the order of 10^{15} cm^{-3} for $Cd_{0.93}Mn_{0.07}Te$ and 10^{16} cm^{-3} for $Cd_{0.9}Mn_{0.1}Te$ samples.

For the photoconductivity measurements ohmic contacts were produced by indium soldering onto the fresh frontside surfaces of the samples. The four point probe method was applied. A Keithley constant current source was used and the voltage drop across the sample was measured at 10 μA constant current. The photoconductivity transients were recorded at 77 K after exposing the samples to monochromatic light of below band-gap energy equal to 1.24 eV ($\lambda = 1.0\ \mu$m). The maximum value of light intensity used in the experiment was of the order of several tens of W m^{-2}. A tungsten lamp served as a light source. A shutter of time constant equal to 0.2 s was applied to turn on and off the light. Prior to each measurement a sample was cooled down in darkness to the liquid nitrogen temperature and subsequently illuminated by a photon flux for several minutes until conductivity saturated. Various photon fluxes were applied. Variation of the photon flux was achieved by changing the current of the lamp supplier. The light beam passed through a monochromator and with the help of fiber optics was focused on a sample mounted in a sample holder. A thermopile was used to measure the photon flux.

4. Results and discussion

In order to analyze the photoconductivity build-up kinetics in terms of the relaxation function $\Phi(t)$ the data have been normalized according to the following formula:

$$\Phi(t) = \Delta\sigma(t) = \frac{\sigma(t_{\mathrm{sat}}) - \sigma(t)}{\sigma(t_{\mathrm{sat}}) - \sigma(t_{\mathrm{on}})}. \tag{7}$$

The normalized photoconductivity transients can be understood as a relaxation function. According to equation (7) $\Delta\sigma(t)$ describes the time evolution of a non-equilibrium state of the investigated physical system and decays monotonically from $\Delta\sigma(t = t_{\mathrm{on}}) = 1$ to $\Delta\sigma(t = t_{\mathrm{sat}}) = 0$ [1]. In the above relation $\Delta\sigma(t)$ denotes relative change in the conductivity due to illumination, $\sigma(t_{\mathrm{on}})$ represents the value of conductivity at the instant of turning the light on and $\sigma(t_{\mathrm{sat}})$ is the saturated conductivity. In figures 1(a) and (b) the relaxation functions are presented for both samples. In both cases the considered kinetics can be well fitted by means of the stretched-exponential relaxation function. In other words, the considered normalized photoconductivity build-up in a time-domain follows the stretched-exponential relaxation pattern.

In figure 2 the dependence of the stretching exponents (determined from the fit of the $\Phi_{\mathrm{KWW}}(t)$ to the photoconductivity build-up kinetics) on a photon flux F is plotted. For both samples the values of α decrease linearly with increasing photon flux. The values of α fall in the range of (0.91–0.99) for $Cd_{0.97}Mn_{0.03}Te$:In whereas for $Cd_{0.9}Mn_{0.1}Te$:In they are smaller and fall in the range of (0.89–0.95). Relaxation time values, resulting from the KWW fitting of the data, were found to be in the range of (350, 120) for $Cd_{0.97}Mn_{0.03}Te$:In and (213, 114) for $Cd_{0.9}Mn_{0.1}Te$:In samples.

4.1. DX centers relaxation rate distribution

As was mentioned in section 2 the explicit form of the relaxation function allows us to gain insight into the local random characteristics of the studied material. The stretched-exponential form of the relaxation function is a consequence of the heavy-tailed distribution of DX center relaxation rates. In figure 3 the densities of the relaxation rates are plotted on a linear and a double-logarithmic scale. The densities were

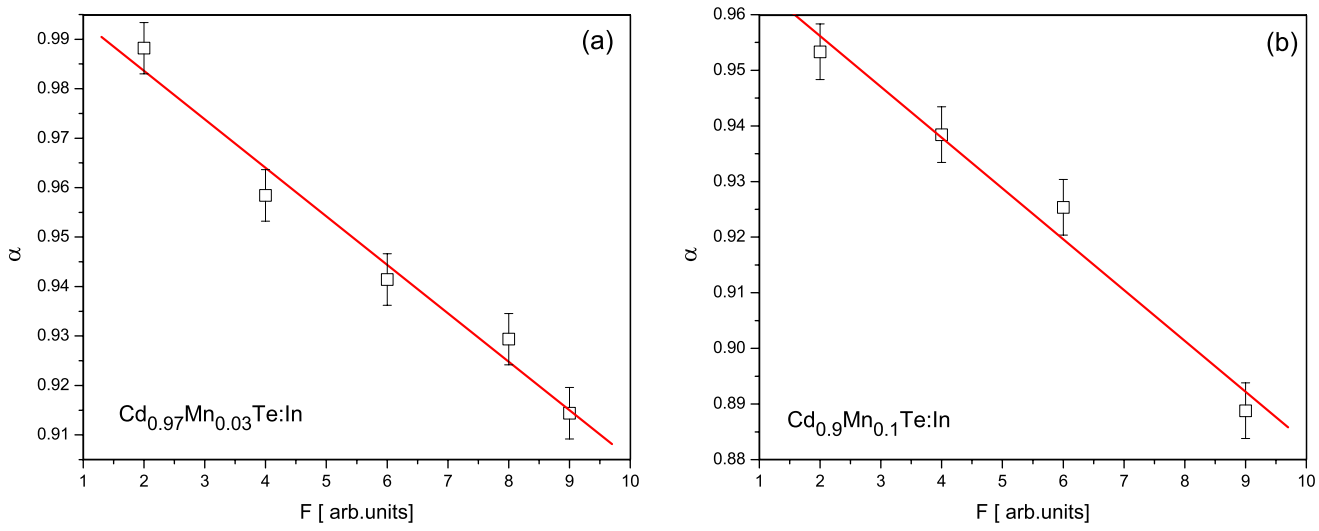

Figure 2. The dependence of the stretching exponent α on the photon flux F.

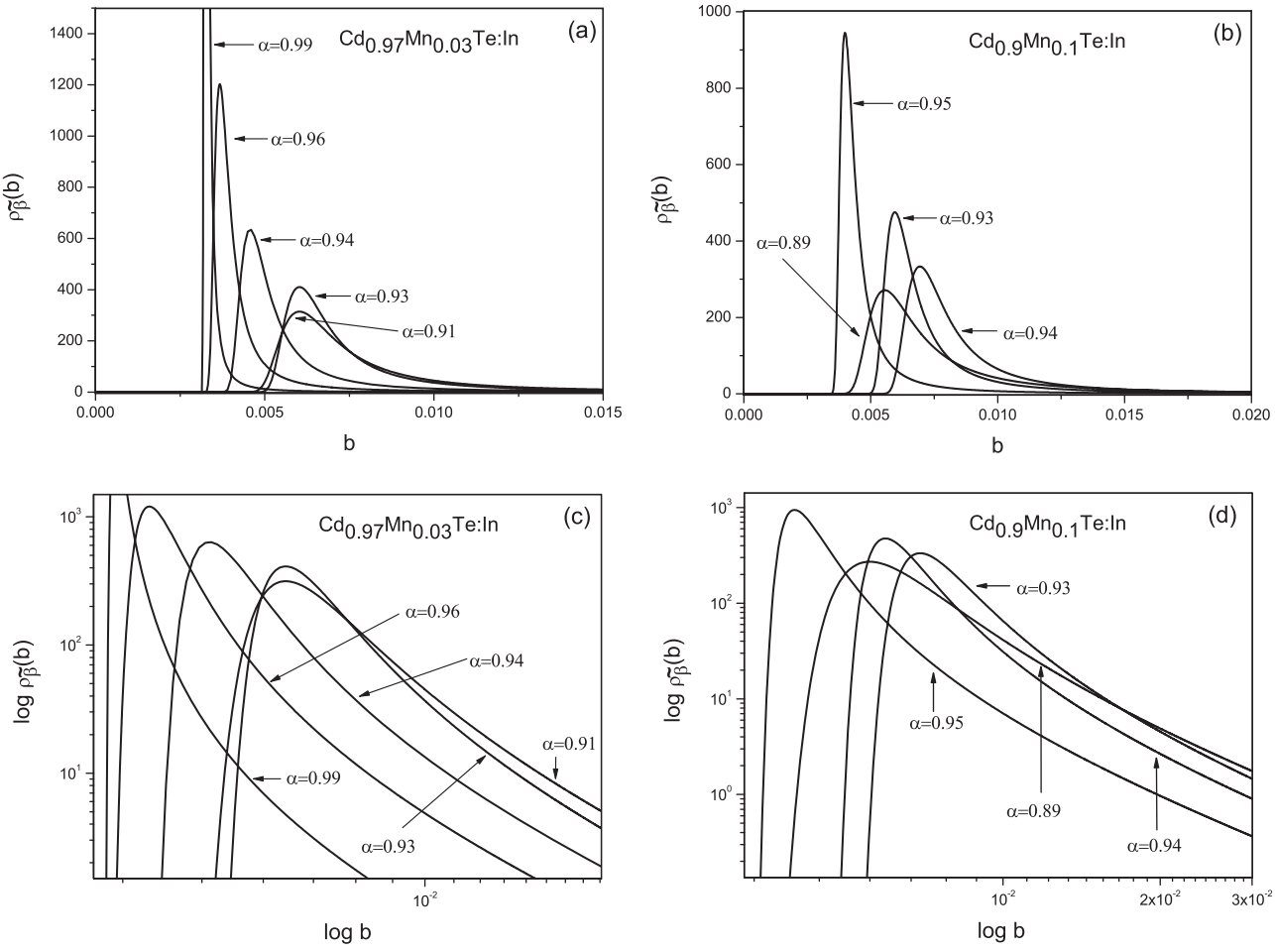

Figure 3. Densities of the relaxation rates for various values of the parameter $\alpha \in (0, 1)$. The upper panels refer to the linear, the lower one the double-logarithmic scale, respectively.

calculated with the use of equation (5) with the values of stretching exponents α and time constants τ resulting from the fitting of the KWW function to the experimental data. The location parameter γ was set to 0. The value of scaling constant c was calculated with use of the formula: $c = [\cos(\frac{\pi\alpha}{2})]^{1/\alpha} \cdot \frac{1}{\tau}$. The obtained densities of the relaxation rates are completely asymmetric ($\beta = 1$). The densities presented in a double-logarithmic scale (figures 3(c) and (d)) demonstrate that the

smaller the stretching parameter is the slower the density decays. Hence the smaller the stretching exponent is the more dispersive the material becomes. Figures 3(a)–(d) lead to two major conclusions.

- increasing photon flux results in the decreasing value of α. This can be explained by the fact that the higher the flux the larger is the number of ionized centers. This means that the probability that the relaxation rates of DX centers will take a value located in the tail of their distribution increases with increasing photon flux.
- comparing figures 3(a) and (b) with figures 3(c) and (d) one can see that the values of α are smaller for the material of higher manganese content. The physical origin of the stretched-exponential response for $Cd_{1-x}Mn_xTe$:Ga has been explained by different local atomic arrangements in this multiternary alloy [14]. If one assumes that this is valid also for indium doped material then actually the higher manganese content leads to the smaller values of α.

According to the stochastic model of relaxation the system responding non-exponentially appears to be divided into clusters. Interactions among the clusters may lead to the creation of some mesoscopic regions ('superclusters'). It can be proven [7, 8, 26] that if the relaxation function takes the KWW form then: (i) the considered system is divided into clusters of similar sizes but of different relaxation rates which may differ by many orders of magnitude, (ii) there is no long-range interactions among relaxing entities. The correlated cluster scheme seems to be consistent with the hypothesis that negatively charged and ionized DX centers interact forming dipole-like DX^-–DX^+ objects [27–31]. It is reasonable to assume that the cluster is formed by such a dipole whereas the interaction among the dipoles results in the appearance of mesoscopic regions—superclusters. Both the interaction inside a cluster and among clusters are presumably short-range interactions and hence the KWW relaxation function is observed in experiment. The fact of the presence of spatial correlations of in-donor charges in CdTe crystals was experimentally confirmed [31]. However, the stochastic hypothesis concerning the presence of short-range correlation between the centers in the studied $Cd_{0.97}Mn_{0.03}Te$:In and $Cd_{0.9}Mn_{0.1}Te$:In should be confirmed by further experimental investigations.

5. Conclusions

The kinetics of photoconductivity build-up due to DX centers was studied in indium doped $Cd_{1-x}Mn_xTe$. It was found that the obtained data can be properly described in terms of the stretched-exponential relaxation pattern. This is due to the fact that the KWW function perfectly fits the short-time power-law exhibited by a response of the transients. It was shown that the distribution of the DX centers' relaxation rates is a heavy-tailed one. A 'weight' of the distribution tail is determined directly by a value of stretching exponent α and indirectly by a photon flux F. Increasing photon flux results in a linear decrease of α. The values of α are lower for higher manganese contents. This observation confirms the assumption that the heavy-tailed distribution of DX centers has its origin in a different local atomic neighborhood in the alloy.

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