Heavily Doped Zinc Oxide with Plasma Frequencies in the Telecommunication Wavelength Range

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1. Introduction

Transparent conductive oxides (TCOs) have been used as transparent electrodes for solar cell applications in the past two decades, but recently also gained attention in plasmonics and nanophotonics due to their low optical loss in the visible, metal-like behavior in the infrared, tailorable optical properties, and well-established fabrication procedures. N-type-doped zinc oxide (ZnO) is especially attractive because its complex permittivity can be engineered over a broad wavelength range by varying the carrier concentration. The carrier concentrations for electronics applications in the semiconductor industry are typically in the range of \(10^{18} - 10^{19} \text{ cm}^{-3}\), corresponding to plasma frequencies in the mid-infrared. To reach plasma wavelengths in the telecom range, carrier concentrations well above \(10^{20} \text{ cm}^{-3}\) are required, which is close to the solid-solubility limit (SSL) of n-type dopants in ZnO. Indeed, it has been demonstrated that ZnO prepared by different deposition techniques, such as magnetron sputtering or pulsed-laser deposition, can have maximum free-carrier concentrations in the range of \((2 - 10) \times 10^{20} \text{ cm}^{-3}\).

The SSL describes the upper limit of the concentration of impurities that are soluble in a solid at thermodynamic equilibrium for a given temperature. Impurity concentrations above the SSL typically lead to the reduction of the free-carrier concentration due to multiple effects, such as secondary phase formation, dopant segregation, out-diffusion, or formation of dimers, which can be electron traps. However, doping beyond the SSL (i.e., hyper-doping) should be feasible via nonequilibrium annealing, which can suppress the processes that reduce the carrier concentration.

To date, there are only a few reports about hyper-doped materials, because nonequilibrium preparation processes are required to overcome the SSL. Hyper-doped silicon was realized by femtosecond laser irradiation in a sulfur and nitrogen atmosphere, which resulted in very rough surfaces with microstructures. Another procedure for hyper-doping is the use of ion implantation and a subsequent short-time annealing process. Ion implantation is a conventional technique to dope materials with nearly any element over a broad doping-level range, but also induces damage next to the incorporation of the dopants. Post-implantation annealing treatments are necessary to remove...
the irradiation damage and to activate the dopants. The use of laser or flash-lamp annealing, which can anneal on the timescale of femtoseconds to microseconds, has enabled arsenic-hyperdoped silicon and gold-hyperdoped germanium.\textsuperscript{19,20}

The SSL of dopants in ZnO strongly depends on the temperature and is rarely reported in the literature. Yoon et al.\textsuperscript{[21]} presented X-ray diffraction (XRD) and Raman measurements on gallium (Ga)-doped ZnO samples that were annealed at 1300 °C for 5 h. They determined an SSL of 0.5 at% (4.2 × 10^20 cm\(^{-3}\)) for this temperature, with higher Ga concentration resulting in the formation of secondary oxide phases.\textsuperscript{[21]} Sky et al.\textsuperscript{[22]} performed isochronal (constant annealing time of 30 min) and isothermal (annealing times between 20 min and 5 h) diffusion experiments for Ga dopants in single-crystalline ZnO in the temperature range of 900–1050 °C and measured the concentration versus depth profiles of Ga using SIMS. Their modeling of the measured Ga diffusion profiles revealed that the dominant diffusion mechanism is consistent with zinc (Zn) vacancy (V\textsubscript{Zn}) mediation via the formation and dissociation of Ga\textsubscript{2}O\textsubscript{3}V\textsubscript{Zn} complexes with substitutional Ga at the Zn sub-lattice (Ga\textsubscript{Zn}).\textsuperscript{[22]} The temperature-dependent SSL of Ga, aluminum (Al), and indium (In) in ZnO was determined by Sky et al. with a reaction-diffusion-type model,\textsuperscript{[22–24]} which is based on Fickian diffusion plus a nonlinear reaction term. More theoretical background can be found in refs.\textsuperscript{[22,23]}

Figure 1a depicts the SSL for the three n-type dopants in ZnO, Ga, Al, and In, as a function of temperature.\textsuperscript{[22–24]} Ga has the highest SSL over a wide temperature range, which makes it the most promising dopant for ZnO plasmonics. This is mainly connected to the small tetrahedral covalent radius and the low formation energy of Ga\textsubscript{2}O\textsubscript{3} compared to those of Al\textsubscript{2}O\textsubscript{3} and In\textsubscript{2}O\textsubscript{3}.\textsuperscript{[13]}

Beyond the choice of the dopant for ion implantation, the annealing time is another key factor. The annealing time must be shorter than or at least similar to the thermal response time (time of a material to react to a sudden temperature change) of ZnO to achieve hyper-doping, so that the dopant diffusion length is less than the average distance between dopants at a given temperature to avoiding clustering.\textsuperscript{[14]} Figure 1b displays the dopant diffusion length \(d_l\) versus the annealing time of the three n-type dopants in ZnO at 900 °C. This diffusion length was calculated using

\[ D = D_0 \times e^{-\left(\frac{E_a}{kT}\right)}, \quad d_l = \sqrt{D \times t_{\text{ann}}} \]  

with the dopant diffusivity \(D\), effective diffusion constant \(D_0\text{eff}\), activation energy \(E_a\), Boltzmann constant \(k\), temperature \(T\), and annealing time \(t_{\text{ann}}\). We extracted the values of \(D_0\text{eff}\) and \(E_a\) for the dopants Ga, Al, and In from references\textsuperscript{[23,25,26]} where they modeled the dopant diffusion profiles at different annealing temperatures. Consequently, we used the values of \(D_0\text{eff}\) of 0.08 cm\(^2\) s\(^{-1}\) and \(E_a\) of 3 ± 0.2 eV for Ga,\textsuperscript{[25]} \(D_0\text{eff}\) = 0.04 cm\(^2\) s\(^{-1}\) and \(E_a\) of 2.6 ± 0.2 eV for Al\textsuperscript{[23]} and \(D_0\text{eff}\) = 0.04 cm\(^2\) s\(^{-1}\) and \(E_a\) of 2.2 ± 0.2 eV for In.\textsuperscript{[26]} The dopant diffusion \(D\) versus annealing time in Figure 1b was then calculated using Equation (1). Long-term annealing procedures, like furnace annealing (FA) with typical annealing times on the order of 10 min, resulting in diffusion lengths of \(d_l(Ga)\) = 280 nm. However, laser annealing (LA) with annealing times in the nanoseconds range strongly suppresses dopant diffusion, resulting in a calculated diffusion length below 0.001 nm, which is smaller than the lattice constants of ZnO. Therefore, we implanted Ga into ZnO and used subsequent laser annealing to achieve heavy doping and plasma wavelengths close to the important telecommunication wavelength of 1.55 μm.

2. Experimental Section

Single-crystalline <0001> ZnO substrates were irradiated with 30 keV Ga\(^+\) ions at room temperature using a commercial focused ion beam (FIB) system. The ZnO substrates were single-side polished and were grown by a hydrothermal process at CrysTec GmbH with a thickness of 0.5 mm and an area of 1 cm\(^2\). Regions with sizes of 150 × 150 μm\(^2\) were homogeneously irradiated. Figure 1c shows implantation profiles for different peak dopant concentrations \(c_p\) as a function of depth, as simulated with the Monte Carlo code TRIM.\textsuperscript{[27]} The projected mean ion range is ≈14 nm and the straggle ≈7 nm. Each sample was implanted with ion fluences ranging from

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure1.png}
\caption{a) Solid-solubility-limit as a function of temperature for n-type dopants (Ga, Al, In) in single-crystalline ZnO, using data taken from references\textsuperscript{[22–24]} b) Dopant diffusion lengths for Ga, Al, and In in ZnO at 900 °C as a function of annealing time\textsuperscript{[23,25,26]} The dashed orange lines indicate typical annealing times for furnace annealing (FA) and laser annealing (LA). c) Simulated Ga implantation profiles in ZnO using SRIM for an ion energy of 30 keV as a function of depth for different ion fluences, resulting in different gallium concentrations \(c_{Ga}\).}
\end{figure}
$6 \times 10^{14}$ to $6 \times 10^{15}$ cm$^{-2}$, resulting in peak values of 0.5 to 5.2 at% that correspond to Ga concentrations of $4.28 \times 10^{20}$ to $4.28 \times 10^{21}$ cm$^{-3}$, respectively.\(^{[28]}\)

Post-implantation annealing was performed with an INNOVAVENT VOLCANO UV laser line beam system, which was powered by a TRUMPF TruMicro 7370 laser with a peak laser power $P_{\text{laser}}$ of 180 W at 10 kHz, with a pulse width of 15 ns. The UV laser light ($\lambda_{\text{laser}} = 343$ nm) was formed into a beam with a “line” cross-section, with full-width at half-maximum (FWHM) of 30 μm with Gaussian shape in the short axis and top-hat profile with a homogeneity better than 5% (pulse peak-to-pulse peak) at a line length of 15 mm in the long axis. This line beam was scanned at a velocity of 30 mm s$^{-1}$ over the sample surface with a 3 μm pitch, resulting in a consecutive laser spot area overlap of 90%. The laser energy density could be set by a variable attenuator to energy densities of 100, 150, 200, 250, 300, and 350 mJ cm$^{-2}$ resulting in different annealing temperature profiles and peak annealing temperatures. The peak annealing temperatures were simulated (see Supporting Information S1) to be about 570 °C (100 mJ cm$^{-2}$), 920 °C (150 mJ cm$^{-2}$), 1300 °C (200 mJ cm$^{-2}$), and 1700 °C (250 mJ cm$^{-2}$). For laser energy densities above 250 μJ cm$^{-2}$, the peak annealing temperatures reach the melting point of ZnO, which is 1975 °C. A first-order phase transition in a material always needs additional latent heat where the temperature does not increase while the phase change is occurring. Increasing the laser energy densities beyond 250 mJ cm$^{-2}$, the extent of melting in ZnO increases and the temperature gradient between heated regions at the surface and the bulk material increases and results in mechanical stress with destructive surface effects, like cracks or partial ablation (see Supporting Information part 2). The onset of these destructive effects sets the maximum laser energy density for successful laser annealing of Ga-doped ZnO to about 250 mJ cm$^{-2}$. Regarding the surface quality of our Ga-doped and laser-annealed ZnO samples, atomic force microscopy (AFM) measurements were performed after the ion implantation and laser annealing. The surface quality was represented by the root mean square (RMS) surface roughness for an area of 2 μm $\times$ 2 μm. A maximum $R_{\text{RMS}}$ value of 1.8 nm was measured for Ga-doped ZnO samples, which were highly doped ($c_p = 5.2$ at%) and annealed at 150–250 mJ cm$^{-2}$. This value is only a factor of seven higher compared to the pristine ZnO, and not orders of magnitude, and thus the surfaces can be still considered of good quality.

The comparison of $R_{\text{RMS}}$ values from our heavily doped ZnO with as-grown doped ZnO thin films grown\(^{[29–31]}\) by different deposition methods shows that our highly doped ZnO samples have a better surface quality for a broad range. For detailed information, please look up Section S4, Supporting Information.

### 3. Results and Discussion

Figure 2(a, top) shows reflectance spectra in the infrared regime obtained from two as-implanted and non-annealed samples with Ga peak concentrations $c_p$ of 0.5 and 5.2 at%, measured using a Fourier-transform infrared (FTIR) spectrometer (Bruker Vertex 70) connected to an infrared microscope (Hyperion 2000) in reflection mode. The spectrum of an intrinsic ZnO substrate is also shown for comparison (black), which is at top of Figure 2a–c similar to the reflectance of the sample with $c_p$ of 0.5 at% (red). The reflectance increased with increasing Ga concentration in the as-implanted state (i.e., before laser annealing). This may be caused by various effects: the formation of Ga clusters at such high impurity concentrations\(^{[32,33]}\) or implanted dopants are in a substitutional position within the ZnO lattice without any annealing, which was experimentally observed for dopants in several references\(^{[34–39]}\).

To clarify the increasing reflectance with increasing Ga concentration in the as-implanted state, we also irradiated, with a conventional ion implanter instead of the FIB, single-crystalline ZnO with Krypton (Kr) at 30 keV and an ion fluence of $4.6 \times 10^{15}$ cm$^{-2}$, to generate a similar defect concentration distribution as for the Ga implantation with $6 \times 10^{15}$ cm$^{-2}$.

![Figure 2](https://example.com/figure2.png)

**Figure 2.** a) Measured (solid lines) and fitted (dotted lines) reflectance spectra of intrinsic (black) and Ga-implanted ZnO (red, blue) in the unannealed state and after laser annealing with different laser energy densities (150 and 250 μJ cm$^{-2}$). b,c) Real part ($n$) and imaginary part ($\kappa$) of the complex refractive index $n = \kappa^n$ extracted from Drude–Lorentz fits to the experimental reflectance spectra from (a) for the gallium-implanted top layer. The perpendicular red dashed line in (b,c) indicates the plasma wavelengths, where $\kappa$ starting to get bigger than $n$ for Ga implanted ZnO with $c_p = 0.5$ at%, which corresponds to plasmonic behavior.
(see Supporting Information of reference[40]). The implanted, chemically inert Kr atoms do not form bonds and do not result in doping. Thus, by comparing the data of both sets of samples, we isolated the influence of implantation-induced defects on the reflectance. The reflectance of the Kr-implanted samples did not change with respect to the increasing ion fluence and is comparable to that of the pristine ZnO sample (see Supporting Information of reference[40]). Thus, we infer that the defects and damage due to ion implantation are not responsible for the strong increasing reflectance in the infrared for the Ga-implanted samples.

The difference in reflectance for Kr and Ga implantation is likely connected to chemical and electronic doping by the implanted Ga, which is not the case for Kr. Gallium acts as a donor on Zn sub-lattice sites, and gallium clusters or charged defect complexes like Zn-GaV_{Ga} could be also formed, which all can increase the free-carrier concentration resulting in an increase in infrared reflectance without annealing. The substitutional position of implanted dopants on Zn-sites in ZnO directly after implantation at room temperature has been found experimentally for many dopants (i.e., Fe,[34] Cu,[35] Ag,[36] Er,[37,38] and Tm[39]) by Rutherford backscattering/channeling spectrometry and emission channeling spectroscopy. Note that, typically, ZnO is not amorphized upon such implantation conditions, with ion fluences lower than 10^{16} \text{cm}^{-2}[41,42].

The optical constants used to describe the intrinsic and unannealed Ga-doped ZnO substrate were extracted from ellipsometry measurements, published in the Supporting Information of reference[40] and also presented for the as-implanted Ga-doped ZnO in the Section S5, Supporting Information. To quantitatively study the optical properties, i.e., the complex refractive index, of the heavily doped ZnO, we built a two-layer optical model to fit our reflectance measurements: a semi-infinite substrate of intrinsic ZnO and a top layer of highly doped ZnO:Ga, for which the optical properties can be well described using Drude-Lorentz oscillations functions[43–47]:

\[
\varepsilon(\omega) = \varepsilon_0 + \varepsilon_\infty \left( 1 - \frac{\omega_p^2}{\omega^2 + i\Gamma_0 \omega} \right) + \Delta \varepsilon_1 \times \left( \frac{\omega_p^2 - i\gamma \omega}{\omega_p^2 - \omega^2 - 2i\kappa\omega} \right) \tag{2}
\]

We used Lorentzian oscillators to model the vibrational modes in the infrared, which we expect to be present in both intrinsic ZnO and in ZnO:Ga. All parameters of the Lorentzian function were taken from references[48–53] and set as constants during fitting. We used the Drude term to estimate the free-electron contribution due to substitutional Ga^{+} doping. We set the screened plasma frequency \(\omega_p\) and damping factor \(\Gamma_0\) of the Drude model to be the fitting parameters, which were swept to find a reflectance that matched with our FTIR measurements. The fitting parameters \(\omega_p\) and \(\Gamma_0\) on the reflectance are crucial for further data processing to calculate the free carrier concentration \(N\), plasma wavelength \(\lambda_p\) and dopant activation efficiency (DAE).

The parameters \(\omega_p\) and \(\omega_N\) are known as frequencies of the plasma resonance and defined as

\[
\omega_p^2 = \frac{Ne^2}{\varepsilon_0 m^*}, \quad \omega_N^2 = \frac{Ne^2}{\varepsilon_\infty \varepsilon_0 m^*} \tag{3}
\]

Note that the relation between \(\omega_p\) and \(\omega_N\) is \(\omega_N^2 = \omega_p^2 / \varepsilon_\infty\), where \(\omega_p\) is named in some literature as screened plasma frequency.

The plasma wavelength \(\lambda_p\) can be calculated with the relation between the energy of electromagnetic radiation and their wavelength

\[
E = \frac{hc}{\lambda}, \quad \lambda_p(\text{nm}) = \frac{1239.83}{E(\text{eV})} \tag{4}
\]

where \(c\) is the speed of light and \(h\) the Planck’s constant. Using Equation (3) and (4), we determined the free carrier concentration \(N\) of our samples. DAE describes the ratio between activated free carrier concentration and the average incorporated dopant concentration for a depth of 30 nm, obtained from the SRIM calculated Gaussian ion implantation profile, see Figure 1c. The average dopant concentration is calculated by an integral over the gallium concentration profile over sample depth and divided by the 30 nm thickness of the Ga-doped ZnO layer. For detailed information see Supporting Information S3 and S5.

For the layer of Ga-implanted ZnO, we assumed a homogeneous film with a thickness of 30 ± 10 nm, which covers 97% of the implanted Ga profile, according to TRIM simulations shown in Figure 1c. This assumption for the thickness is also well justified by the anticipated lack of diffusion during the annealing process as shown in Figure 2a, the good agreement between our models (dotted lines) and reflectance measurements (solid curves) enabled us to extract the real and imaginary parts of the complex refractive index of the resulting ZnO:Ga (Figure 2b,c).

A characteristic of Drude-metal-like behavior for a material is when the extinction coefficient \(\Im(\tilde{n}) = n\) becomes higher than the real part of the refractive index \(\Re(\tilde{n}) = n\), i.e., \(\kappa > n\)[54] like in Figure 2b,c this is highlighted with a perpendicular red dashed line, which indicates that Ga-implanted ZnO has a gallium concentration peak \(c_\text{p}\) of 0.5 at%. At this point, we get a plasmonic material, which could be useful for plasmonic applications in the near-infrared.

We performed laser annealing at a laser energy density of 100 mJ cm\(^{-2}\), but did not observe any significant changes in the reflectance signal compared to the as-implanted state, see Supporting Information Information S2. However, reflectance starts to slightly increase at a laser energy density of 150 mJ cm\(^{-2}\) due to dopant activation (Figure 2a). This dopant activation appears to be more effective for low gallium ion fluences. However, stable defect clusters are formed for higher ion fluences, which require higher annealing temperatures for healing.[29,30] An increase in the laser power results in a further increase in the reflectance. For the investigated laser energy densities of 100–350 mJ cm\(^{-2}\), the maximum reflectance signal is achieved using laser energy densities of 250 mJ cm\(^{-2}\), which is shown at the bottom of Figure 2a. This increase can be attributed to the electrical and optical activation of Ga dopants in the ZnO lattice, thus the free-carrier concentration increases. A further increase of the laser energy density to 300 mJ cm\(^{-2}\) and
Figure 3. a) Plasma frequency $\omega_N$ and b) damping factor $\Gamma_D$ extracted from the fits shown in Figure 2 as a function of the ion fluences for different laser energy densities. c) Dopant activation efficiency, which is the ratio of activated free carriers to the averaged implanted Ga concentration across the thickness, versus ion fluences. d) Plasma wavelength $\lambda_p$ versus ion fluence. The telecommunication wavelength is indicated with a blue dashed line.

above resulted in the degradation of the sample, which we observed with a strong increase in the reflectance of our intrinsic ZnO reference sample. The reflectance increase is caused by the sublimation of the surface layer, where the oxygen diffuses out and a Zn-enriched surface layer forms. Via SEM, we investigated cracks through the ZnO and partly ablated portions of the material, see Supporting Information part S2.

Figure 3a,b shows the plasma frequency $\omega_N$ and damping factor $\Gamma_D$ as a function of the ion fluence, where $\omega_N$ and $\Gamma_D$ were extracted from the fits to reflectance spectra in Figure 2. Details and the complete data sets are present in Supporting Information S2. Increasing both the ion fluence and the laser energy density usually leads to an increase of $\omega_N$ and the highest plasma frequencies of 1–1.3 eV were obtained for a laser energy density of 250 mJ cm$^{-2}$ and an ion fluence range of $3.6 \times 10^{15}$–$6 \times 10^{15}$ cm$^{-2}$ (equivalently, peak dopant concentration of 3.1–5.2 at%). Degradation of the ZnO samples occurs beyond this value, as mentioned earlier. The damping factor is inversely proportional to the carrier mobility and Figure 3b indicates that higher dopant concentrations result in a lower mobility, which is consistent with increased electron-defect scattering.

Figure 3c depicts the dopant activation efficiency, which is the ratio of the achieved free-carrier concentration for a given averaged implanted Ga concentration across the thickness of 30 nm. Laser annealing with 150 mJ cm$^{-2}$ results in activation of 95% to 40% of the incorporated Ga atoms with increasing ion fluence, whereas values of 100%–50% can be achieved by laser annealing using 250 mJ cm$^{-2}$. However, the activation efficiency also decreases with increasing dopant concentration, which is most probably limited by the temperature-dependent SSL of Ga in ZnO.

Furthermore, we extracted the activated dopant concentration from the fits and plotted in Figure 3d the respective plasma wavelength, versus the used ion fluence range for all investigated ZnO:Ga layers. We obtained the highest carrier concentration of $(1.23 \pm 0.41) \times 10^{21}$ cm$^{-3}$ for an ion fluence of $6 \times 10^{15}$ cm$^{-2}$ and for laser annealing with a laser energy density of 250 mJ cm$^{-2}$. This value corresponds to a plasma wavelength of $1.02 \pm 0.17 \mu m$, which is below the telecommunication wavelength of 1.55 µm (blue dashed line in Figure 3d). This value translates also to an activation of $\approx 10^{21}$ cm$^{-3}$ Ga atoms in the ZnO lattice, which is close to the SSL of Ga at a temperature of 1975°C (compare Figure 1a).

Figure 4 highlights the SSL of Ga compared to the activated dopant concentrations of our ZnO:Ga samples, irradiated with an ion fluence of $6 \times 10^{15}$ cm$^{-2}$ ($c_p = 5.2$ at%), for the investigated temperature range. Here, the laser energy has been converted to a temperature using the material parameters of ZnO, the Beer–Lambert law, and the heat equation (see Supporting Information S1). For the laser energy density range of 150–250 mJ cm$^{-2}$, we achieved hyper-doped ZnO. Increasing the laser energy density toward the melting temperature of ZnO led to values close to the SSL of Ga in ZnO.$^{[22]}$

4. Conclusion

In conclusion, we gradually tuned the optical properties of ZnO by ion implantation of gallium and subsequent nanosecond laser annealing. Using this approach, we achieved even plasma wavelengths in the near-infrared spectral region, including for wavelengths shorter than 1.55 µm. The highest achieved free-carrier concentration in ZnO:Ga was $N_c = 1.23 \pm 0.41 \times 10^{21}$ cm$^{-3}$, corresponding to $\lambda_p = 1.02 \pm 0.17 \mu m$, which is 1.48 at% substituted Gallium in the ZnO host lattice with a doping level of $2.16 \times 10^{23}$ cm$^{-3}$ (2.6 at%) and a dopant activation efficiency
of 57%. In general, with this doping approach, we are reaching dopant activation efficiencies in the range of 30% up to 100%. This is enabled by the combination of high-concentration ion implantation and the high peak annealing temperatures during the short laser annealing in the nanosecond regime, where dopant diffusion is suppressed, which is advantageous for maintaining spatially localized distributions of dopants, resulting even in hyper-doping of ZnO under certain conditions. Heavily or hyper-doped ZnO:Ga with plasma frequencies in the telecommunication range have a broad applicability. With an FIB, we are able to selectively and gradually dope ZnO. Thus, it opens a wide field for lateral arrangements of sub-wavelength sized and spaced optical doped and un-doped regions with a high optical contrast, which is a promising approach for photonic or plasmonic applications in the NIR spectral range, for example with an area selective ion implantation process.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

hyper doping, ion implantation, laser annealing, non-equilibrium, plasmonics

Figure 4. Comparison between activated free-carrier concentrations, from the Ga-doped samples irradiated with an ion fluence of $6 \times 10^{15}$ cm$^{-2}$ ($c_0 = 5.2$ at%) and laser annealed in the laser energy density range 150–250 mJ cm$^{-2}$ (corresponds to an annealing temperature range 920 and 1700°C) and the temperature-dependent solid solubility limit of Ga in ZnO taken from the work of Sky et al.[22] where the gray area around the black line displays an error of ±18% for the solid solubility limit.
