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Self-diffusion in Germanium-rich liquid Germanium-Nickel investigated by quasielastic neutron scattering

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Abstract

We report atomic dynamics in liquid $Ge_{98}Ni_2$ measured using quasielastic neutron scattering. Isotopic substitution enabled to separately determine Ge and Ni self-diffusion with high accuracy. The Ge self-diffusion coefficient in liquid $Ge_{98}Ni_2$ is equal within error bars to that in pure liquid Ge. However, the Ni self-diffusion coefficient lies at least 30% below the Ge self-diffusion coefficient. This behaviour differs from previously reported atomic dynamics in Ge-rich GeAu, GeSi, GeIn and GeCe melts, where no separation of the self-diffusion coefficients between the minor component and Ge is observed. The change of the atomic dynamics already at an addition of 2 at% Ni points to electronic and chemical origins in $Ge_{98}Ni_2$. Moreover, the slower self-diffusion of the minor component compared to Ge might be associated with two different local structural environments, as observed in Ge-Ni alloys at higher Ni concentration.

1. Introduction

Metal impurities are of great concern in semiconductor technologies, both during the fabrication processes and during their applications as integrated circuit devices [1]. Confining incorporation of metal impurities is crucial for the electrical performance and lifetime of these devices. Purification of silicon and Germanium using directional solidification relies on the partitioning of impurities between solid and liquid phases. Modelling of these processes requires understanding of the diffusion process of the metals in both solid and melt, among which transition metal elements like Ni are one of the dominant species [2].

Self-diffusion of a minor additional component is referred to as impurity-diffusion. In solid Ge the self-diffusion coefficients of different metal impurities differ by several orders of magnitude [3] as a result of a vacancy mediated diffusion mechanisms. Upon melting Ge undergoes a semiconductor to metal transition. Electric conductivity in liquid Ge is comparable to liquid metals [4, 5] and density increases by approximately 5% upon melting. However, as opposed to liquid metals, the coordination number in liquid Ge is still low with a value of approximately 6 [6, 7] compared to a value of approximately 12 for liquid Ni, Fe, and Zr [8]. Impurity-diffusion in such loosely packed liquid metals is often modelled by a transport mechanism of hard-sphere like binary collision (see for instance [9] for liquid Sn). Here the diffusion coefficient is expected to depend on atomic mass.

However, atomic mobility in dilute Ge-based alloys reported for $Ge_{98}Si_2$, $Ge_{98}Au_2$, $Ge_{98}Gd_2$, and $Ge_{98}Ce_2$, shows no dependence on the atomic mass of the solute [10]. Moreover, in $Ge_{98}Au_2$, $Ge_{98}In_2$, $Ge_{98}Ce_2$, and $Ge_{98}Si_2$ [6] both the self-diffusion coefficient of Ge and of the impurity atoms diffusion were determined. The resulting diffusion-coefficients are equal within error bars. Results indicate a highly collective atomic transport mechanism which is entirely different compared to that in the solid. These findings are in line with molecular dynamic simulations indicating a microscopic cage effect in liquid Ge [11]. Noteworthy for the larger solutes (Gd and Ce) an overall lower mobility was observed.

Concerning Ni as a diffusion species, in liquid Si which exhibits a similar topological structure compared to liquid Ge [12], the Ni self-diffusion coefficients measured by quasielastic neutron scattering (QNS) in silicon-rich liquid Si-Ni show only a very weak dependence with Ni concentration [13]. However they are almost an order of magnitude faster than in pure liquid Ni [13, 14]. When compared to silicon self-diffusion coefficients using the Stokes-Einstein relation the results agree within the experimental uncertainty [15], indicating a strong correlation between the mobility of the constituents. Similar behaviour has been recently observed for the Ge-rich $Ge_{66.7}Ni_{33.3}$ alloy melt as well [16].

In the following we present diffusion-coefficients in liquid $Ge_{98}Ni_2$ which further advances the understanding of impurity-diffusion in loosely packed liquid metals like Ge. We show that the Ge self-diffusion is equal to the Ge self-diffusion in pure liquid Germanium. The Ni self-diffusion coefficient, on the other hand, is significantly lower compared to the Ge self-diffusion coefficient. This difference in the mass transport behaviour indicates more complex liquid dynamics compared to the systems reported above, which may be related to chemical bonding between Ge and Ni.

2. Experimental details

Samples used for neutron scattering were prepared from high purity ^{nat}Ge purchased from PPM Pure Metals GmbH (99.9999%), ^{nat}Ni purchased from ChemPur GmbH (99.99%), and ^{60}Ni with a 99.6% isotope enrichment and metal impurities in the ppm range purchased from STB Isotope Germany GmbH. Alloys were prepared by arc melting the respective constituents under high purity argon atmosphere. Subsequently the alloys were filled in thin-walled Al_2O_3 crucible (0.5 mm wall thickness), giving a cylindrical sample shape of 9 mm in diameter and 40 mm in height.

Diffusion in liquid $Ge_{98}{}^{nat}Ni_2$ was measured on the neutron time-of-flight spectrometer ToFToF at the Forschungsneutronenquelle Heinz Maier-Leibnitz. An incoming neutron wavelength of 7 Å provided an accessible wavenumber range of 0.3 to 2.0 Å^{-1} at zero energy transfer with an energy resolution of approximately 50 μeV full width at half maximum. Measurements were performed inside a standard niobium resistance furnace. The Al_2O_3 crucibles containing the samples were attached to a niobium holder, and placed in the furnace in the way that only the part of the crucible containing the sample is illuminated by the neutron beam. Measurements were performed at a temperate of 1273 K which is slightly above the melting point of Ge $T_m = 1211$ K.

Quasielastic neutron scattering on liquid $Ge_{98}^{60}Ni_2$ was performed on the crystal time-of-flight spectrometer IN6 at the Institut Laue-Langevin using the same sample geometry. An incoming neutron wavelength of 5.1 Å^{-1} gave an accessible wavenumber range of 0.4 to 2.0 Å^{-1} at zero energy transfer with an instrumental energy resolution of about 70 μeV full width at half maximum. A similar niobium resistance furnace was used as the one described above. The sample was processed at temperatures of 1273 K and 1520 K.

For both experiments the sample was processed under a vacuum below 1×10^{-5} mbar. No measurable loss of sample mass due to evaporation was identified. The Al_2O_3 crucible is inert to the GeNi melt at the investigated temperatures, and its incoherent scattering contribution to the signal was negligible. Experiments were performed similarly to previously reported (see also [6, 10, 17]).

All neutron scattering spectra were normalised to a vanadium standard, corrected for self-absorption and empty container scattering and interpolated to constant wave number q to obtain the dynamic structure factor $S(q, \omega)$. All spectra were fitted by a Lorentzian function:

$$S(q,\,\omega) = \frac{A}{\pi}\frac{\Gamma(q)}{(\hbar\omega^2) + (\Gamma(q))^2} \otimes R(q,\,\omega) + b(q) \tag{1}$$

convoluted with the instrumental resolution function $R(q, \omega)$. Here, $b(q)$ denotes an energy independent background and $\Gamma(q)$ the half width at half maximum of the Lorentzian curve (see figure 1). The methodology is the same as previously reported [10, 17].

In general, the dynamic structure factor $S(q, \omega)$ equals the sum of a coherent and incoherent part. As demonstrated in [17] for small q and within the observed energy range, in monoatomic liquids the quasielastic line in $S(q, \omega)$ originates from incoherent scattering in case of an existing incoherent scattering cross section. For alloys, coherent contributions due to concentration-concentration fluctuation (S_{cc}) may contribute to the quasielastic line. However, in our case even with the slightly higher coherent contributions in the case of the $Ge_{98}^{60}Ni_2$ compared to the $Ge_{98}^{nat}Ni_2$ sample, the impact of interdiffusion to the scattering signal can be neglected (see appendix). Self-diffusion of the individual components can therefore be accurately determined.

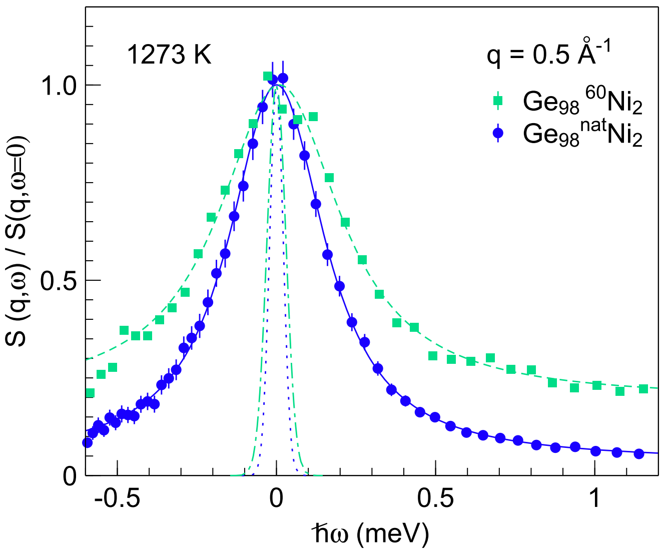

Figure 1. Measured dynamic structure factors of $Ge_{98}^{60}Ni_2$ (filled squares) and $Ge_{98}^{nat}Ni_2$ (full dots) at 1273 K. Dashed and solid lines are fits using equation (1), respectively. The dotted and dash-dotted lines represent the instrumental energy resolution functions at the TofTof and at the IN6 spectrometers, respectively, where the instrumental energy resolution curve of IN6 is slightly broader. Measured spectra are normalized to the respective $S(q, \omega = 0)$ values. For this reason the background appears higher in case of $Ge_{98}^{60}Ni_2$. The $b(q)$ obtained from the fit of equation (1) are equal within uncertainties.

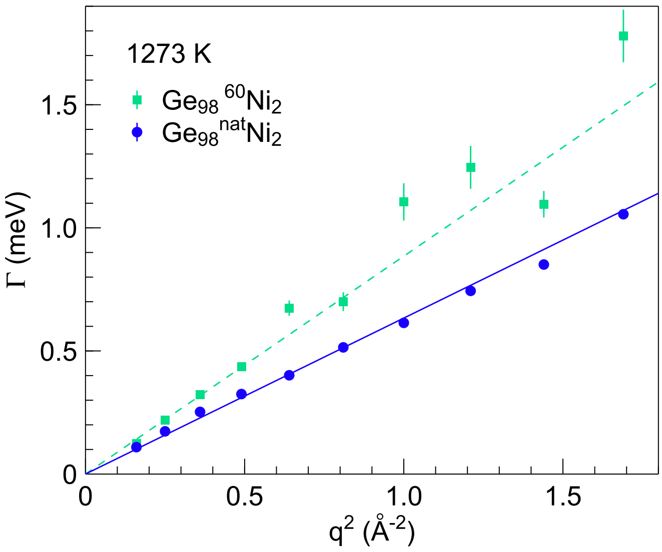

Figure 2. Obtained half width at half maximum $\Gamma(q)$ as a function of q^2 for the $Ge_{98}^{60}Ni_2$ and $Ge_{98}^{nat}Ni_2$ samples at 1273 K, from which self-diffusion coefficients are derived. Legends are the same as in figure 1.

For the $Ge_{98}Ni_2$ samples relative contributions of Ge- and Ni-diffusion to the incoherent scattering signal depend on isotope composition [18]. ^{nat}Ge exhibits an incoherent neutron scattering cross section σ_{inc} of 0.18 barn, ^{nat}Ni of $\sigma_{inc} = 5.2$ barn, and ^{60}Ni of $\sigma_{inc} = 0$ barn. In case chemical incoherence would affect the diffusion coefficient, its contribution for $Ge_{98}^{nat}Ni_2$ respectively for $Ge_{98}^{60}Ni_2$ can be neglected because the concentration of the alloyed Ni is very low, see also [19]. Thus for $Ge_{98}^{60}Ni_2$ only Ge contributes to the incoherent scattering signal and the Ge self-diffusion coefficient may be derived via [20]

$$D_s = \frac{\Gamma(q)}{\hbar q^2}, \tag{2}$$

as can be seen in figure 2. For $Ge_{98}^{nat}Ni_2$, on the other hand, Ge contributes only approximately 60%, and Ni contributes 40% to the incoherent scattering signal. In this case, the diffusion coefficient derived via equation (2) is a weighted average according to the respective scattering contributions.

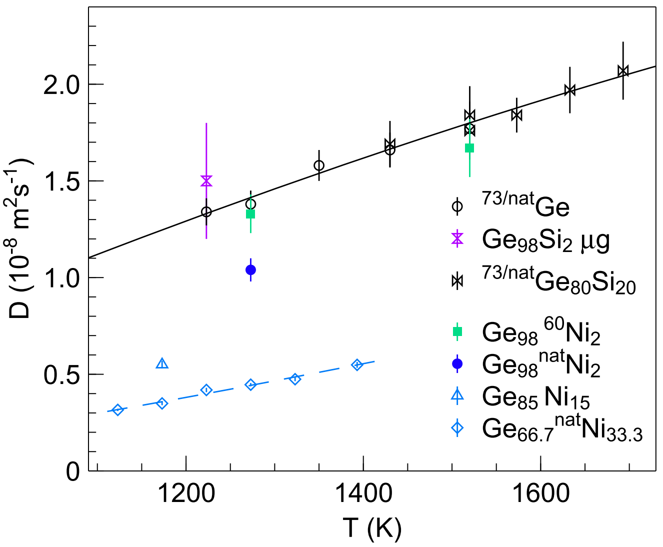

Figure 3. Measured self-diffusion coefficient in Ge-Ni and Ge-Si melts. For liquid $Ge_{98}^{60}Ni_2$ the Ge self-diffusion is measured. For $Ge_{98}^{nat}Ni_2$ the measured self-diffusion coefficient represents a weighted mean diffusion coefficient according to the incoherent contribution of Ge and Ni (60% for Ge and 40% for Ni). For $Ge_{85}Ni_{15}$ and $Ge_{66.7}Ni_{33.3}$ the reported results correspond to Ni self-diffusion [16, 21]. The Ge self-diffusion coefficient in pure Ge, $Ge_{98}Si_2$, $Ge_{80}Si_{20}$ and $Ge_{98}^{60}Ni_2$ agrees within error bars [6, 10].

3. Results and discussion

In figure 3 the diffusion coefficients in liquid $Ge_{98}Ni_2$ derived via QNS are depicted as a function of temperature. In addition, Ge self-diffusion coefficients in pure liquid Ge, $Ge_{80}Si_{20}$, [6, 10] as well as Ni self-diffusion coefficient in liquid $Ge_{85}Ni_{15}$ [21] and $Ge_{66.7}Ni_{33.3}$ [16] are shown for comparison.

The Ge self-diffusion coefficient in pure liquid Ge and the Ge self-diffusion coefficient in liquid $Ge_{98}Ni_2$ (measured using $Ge_{98}^{60}Ni_2$) are equal within error limits, both, at 1273 K and at 1520 K. This in turn indicates a similar temperature dependence in the temperature range between the melting point of Ge and approximately 300 K above. In contrast, the diffusion coefficient measured on the liquid $Ge_{98}^{nat}Ni_2$ sample at 1273 K is $(1.0 \pm 0.1) \times 10^{-8}$ m^{2}s^{-1}, which lies approximately 30% below the Ge self-diffusion coefficient measured using $Ge_{98}^{60}Ni_2$ at the same temperature ($(1.3 \pm 0.1)\times10^{-8}$ m^{2}s^{-1}). Keeping in mind that the measured self-diffusion coefficient in the case of $Ge_{98}^{nat}Ni_2$ represents a weighted mean diffusion coefficient according to the incoherent contribution of Ge and Ni (60% for Ge and 40% for Ni), the Ni self-diffusion coefficient (that is the impurity-diffusion coefficient) in liquid $Ge_{98}Ni_2$ lies at least 30% below the Ge self-diffusion coefficient.

Furthermore, assuming that the Ge self-diffusion coefficient equals the diffusion-coefficient in pure liquid Ge, we additionally fitted the $S(q, \omega)$ spectra of the $Ge_{98}^{nat}Ni_2$ with two Lorentzial lines: one representing the Ge contribution with a fixed diffusion coefficient of $(1.3 \pm 0.1) \times 10^{-8}$ m^{2}s^{-1} (derived from the measured diffusion coefficient in $Ge_{98}^{60}Ni_2$) and one representing Ni diffusion with a variable half width. The resulting Ni self-diffusion coefficient of $(0.7 \pm 0.1) \times 10^{-8}$ m^{2}s^{-1} lies approximately 50% below the Ge self-diffusion coefficient, further supporting the observation that the Ni self-diffusion coefficient lies at least 30% below the Ge self-diffusion coefficient[6].

The Ni self-diffusion coefficient in liquid $Ge_{85}Ni_{15}$ reported by Neumann *et al* [21] is even smaller, on the order of 0.5×10^{-8} m^{2}s^{-1} at 1173 K. Overall the atomic mobility decreases with increasing Ni concentration, as can be seen when compared to the Ni self-diffusion coefficient of $Ge_{66.7}Ni_{33.3}$ (for $Ge_{66.7}Ni_{33.3}$ the Ni concentration is high enough that the incoherent scattering contribution of Ni is dominating, i.e. 93% of the incoherent scattering contribution) [16]. The self-diffusion coefficient of pure liquid Ni at 1514 K is of $(2.09\pm 0.08)\times10^{-9}$ m^{2}s^{-1}, i.e. smaller by a factor of almost 8 [22].

In contrast to Ni, alloying Si to the Ge melt does not affect the melt dynamics significantly. The Ge self-diffusion coefficient in pure liquid Ge measured with QNS and the impurity-diffusion coefficient of Si measured in liquid $Ge_{98}Si_2$ in a long capillary experiment under microgravity conditions are equal within the experimental uncertainties [10]. Also, the Ge self-diffusion coefficient in the $Ge_{80}Si_{20}$ melt does not show measurable changes compared to that in pure Ge [6]. The temperature dependence can be well described by an Arrhenius law with an activation energy of 164 ± 8 meV and a pre-exponential factor of $D_0 = (6.2 \pm 0.4) \times 10^{-8}$ m^{2}s^{-1}.

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[6] Note that for a signal comprising of only Ni self-correlation further isotope substitution measurements may be necessary, for example using ^{75}Ge with zero incoherent scattering signal.

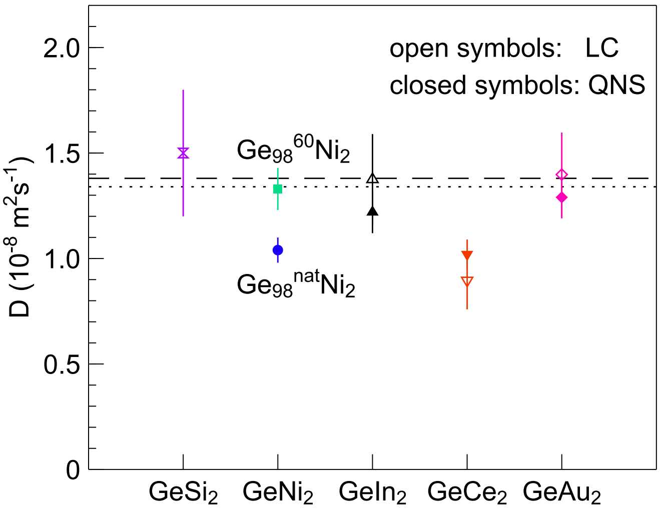

Figure 4. Measured self-diffusion coefficient in dilute $Ge_{98}X_2$ (X = Si, Ni, Au, In, Ce) melts, compared with the Ge self-diffusion coefficient in liquid Germanium. Except for $Ge_{98}Ni_2$, the diffusion coefficient are taken from [6, 10] (open symbols QNS, closed symbols long-capillary), which were measured at 1233 K. The *x*-axis is sorted according to atomic mass of the impurity atoms (Si:28.1 u, Ni: 58.7 u, In: 114.8 u, Ce: 140.1 u, and Au: 197.0 u). For $GeNi_2$ the self-diffusion coefficient measured on $Ge_{98}^{60}Ni_2$ sample corresponds to the Ge self-diffusion coefficient, whereas the self-diffusion coefficient measured on the $Ge_{98}^{60}Ni_2$ sample corresponds to a weighted mean diffusion coefficient of Ge and Ni self-diffusion (s. Figure 3). The dotted and dashed lines are the Ge self-diffusion coefficient at 1233 and 1273 K, respectively, which shows that the change with respect to temperature is within the uncertainty of impurity diffusion coefficients. Only in $Ge_{98}Ni_2$ a separation of individual self-diffusion coefficients can be observed.

In figure 4 the Ge and Ni self-diffusion coefficients determined in this work are compared with the Ge self-diffusion coefficients in liquid $Ge_{98}X_2$ (X = Si, Au, In, Ce) measured using QNS and, if available, with the self-diffusion coefficient of the respective impurity component (measured using capillary experiments). Atomic masses of the studied impurity atoms range from 28.1 u (Si) to 197 u (Au). In case of $Ge_{98}Ni_2$, the atomic mass of Ge (72.6 u) is slightly higher than for Ni (58.7 u). Kinetic theory theory predicts diffusion coefficients to be proportional to the inverse square root of atomic masses. However, a clear dependence of the atomic mobility on atomic mass is not observed, indicating that diffusion may not be described within kinetic theory of hard-sphere like binary collisions. In addition, the observed difference cannot be understood by atomic size as the covalent radius of Ge is almost equal to that of Ni (Ge: 1.2 Å, Ni: 1.24 Å).

For $Ge_{98}Au_2$, $Ge_{98}In_2$, and $Ge_{98}Ce_2$, the Ge self-diffusion coefficient (filled symbols in figure 4), and the self-diffusion coefficients of impurity atoms measured in long-capillary experiments (open symbols in figure 4) [10] are equal within error bars. Likewise, in Ge-rich GeSi Ge- and Si self-diffusion coefficient are equal within error bars [6, 10]. Only in the Ge-rich GeNi alloy reported here, as opposed to Ge-rich GeSi, GeIn, GeCe and GeAu, a separation between the self-diffusion coefficient of Ge and of the impurity atom Ni is observed. Notably, the addition of small amounts of Ce also slows down the overall dynamics of the $Ge_{98}Ce_2$ system by up to a factor of 2. The same behaviours was reported for $Ga_{98}Ni_2$, where addition of small amounts of Ni slows down the overall dynamics [19], The change of atomic dynamics at such low concentration modifications points to an electronic- and chemical origin, similar to the case of alloying Al to Zr-based glass-forming melts [23].

Yet, in contrast to $Ge_{98}Ce_2$ and $Ga_{98}Ni_2$, in $Ge_{98}Ni_2$ only the self-diffusion coefficient of the impurity atom is reduced and not the self-diffusion of Ge. The separation of Ge- and Ni self-diffusion coefficients is expected to come along with structural differences. In fact, structural studies of $Ge_{66.7}Ni_{33.3}$ and $Ge_{80}Ni_{20}$ melt indicate preferred formation of Ge-Ni heterogeneous pairs in the melt [24, 25]. It is even assumed that in $Ge_{66.7}Ni_{33.3}$ there are two different structural environments: one with preferred Ge-Ni pairs showing electronic charge transfer and a second one of the remaining Ge atoms similar to that in the pure Ge melt. These features could explain the liquid dynamics observed here: a local structural environment for Ge atoms in GeNi similar to that in pure liquid Ge would go along with a self-diffusion coefficient similar to the one in pure liquid Ge. For the Ni impurity atoms preferred Ge-Ni pairs may cause a lower mobility of the Ni atoms. In contrast, in Ge-rich GeSi, where no difference in the Ge and Si self-diffusion coefficients is observed, the topological structure of the melt is also very similar to pure Ge [6, 10].

4. Conclusion

We present self-diffusion coefficients in liquid $Ge_{98}Ni_2$ measured using QNS. Isotopic substitution enabled to separately determine Ge- and Ni-diffusion. The Ge self-diffusion coefficient is equal to the Ge self-diffusion in pure liquid Ge. However, Ni self-diffusion lies at least 30% below the Ge self-diffusion coefficient. The atomic mass of Ge (72.6 u) is slightly higher than for Ni (58.7 u). Therefore, as reported for Ge-rich GeAu, GeSi, GeIn and GeCe as well, no clear dependence on atomic mass can be observed, and the diffusion mechanism cannot be described by binary collisions. Except in the case of GeNi, diffusion of the individual components is equal to one another. For the separation of Ge- and Ni self-diffusion as well as the slower impurity atom dynamics in $Ge_{98}Ni_2$, electronic and chemical effects are expected to play an important role. Such effects may lead to two different local structural environments, as shown for Ge-rich GeNi with higher Ni-concentration. However, whether these structural peculiarities in Ge-rich GeNi are also present in the investigated $Ge_{98}Ni_2$ melt will require further structural investigation.

Acknowledgments

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Data availability statement

The data cannot be made publicly available upon publication because the cost of preparing, depositing and hosting the data would be prohibitive within the terms of this research project. The data that support the findings of this study are available upon reasonable request from the authors.

Appendix

The scattering intensity measured in a quasielastic neutron scattering can be divided into a coherent I^{coh} and an incoherent I^{inc} part, both as a function of momentum and energy transfers [26]:

$$I(q, E) = I^{\mathrm{coh}}(q, E) + I^{\mathrm{inc}}(q, E). \tag{A1}$$

While the incoherent contribution is related to incoherent scattering cross sections and is used to derive the self-diffusion coefficient, the coherent part can be expressed, particularly in the case of binary alloys, using the Bhatia-Thornton formalism of partial structure factors by [27]

$$\begin{aligned} I^{\mathrm{coh}}(q, E) = 4\pi[\langle b\rangle^2 S_{NN}(q, E) + 2\langle b\rangle \Delta b S_{NC}(q, E) \\ + c_A c_B (\Delta b)^2 S_{CC}(q, E)], \end{aligned} \tag{A2}$$

where in our case of Ge-Ni $\langle b\rangle = c_{\mathrm{Ni}} b_{\mathrm{Ni}} + c_{\mathrm{Ge}} b_{\mathrm{Ge}}$ and $\Delta b = b_{\mathrm{Ni}} - b_{\mathrm{Ge}}$. $S_{NN}(q, E)$ is the number-number partial structure factor, $S_{NC}(q, E)$ is the number-concatenation partial structure factor, and $S_{CC}(q, E)$ is concentration-concentration partial structure factor (by definition for $q \to \infty$ $S_{CC} \doteq 1$). Normalized to the scattering cross section $\sigma = 4\pi\langle b^2\rangle$, the prefactors of the three partial structure factors are listed in table A1. In both cases the weighting factor of the S_{CC} is small.

At small q, both $S_{NN}(q, E)$ and $S_{NC}(q, E)$ approach a very small value ($S_{NN}(0)$ is related to the isothermal compressibility for a monoatomic system) [20]. The remaining contribution of $S_{CC}(q, E)$ in the hydrodynamic limit $q \to 0$ represents the decay of concentration-concentration correlation. This is related to the interdiffusion, leading to a Lorentzian line shape

$$S_{CC}(q, E) = S_{CC}(q = 0)\frac{\Gamma/2\pi}{E^2 + (\Gamma/2)^2} \text{ for } q \to 0,\ E \to 0. \tag{A3}$$

The full width at half maximum (FWHM) Γ can be expressed as

$$\Gamma = 2\hbar q^2 D^{\mathrm{int}}, \tag{A4}$$

where D^{int} is the interdiffusion constant [28].

Table A1. Normalized prefactors of the contribution from the Bhatia-Thornton partial structure factors S_{NN}, S_{NC}, and S_{CC} for $GeNi_2$.

composition	S_{NN} $\langle b\rangle^2/\langle b^2\rangle$	S_{NC} $2\langle b\rangle\Delta b/\langle b^2\rangle$	S_{CC} $c_{\mathrm{Ge}}c_{\mathrm{Ni}}(\Delta b)^2/\langle b^2\rangle$
$Ge^{nat}_{98}Ni_2$	0.999	0.513	0.001
$Ge^{60}_{98}Ni_2$	0.855	1.322	0.009

In case of diluted alloys, Darken equation (equation (A5)) can be invoked as a good approximation [29], and the thermodynamic factor Φ can be assumed as 1. The interdiffusion coefficient can be given by

$$D^{\text{int}}_{\text{Darken}} = \Phi(c_{\text{Ni}} D_{\text{Ge}} + c_{\text{Ge}} D_{\text{Ni}}), \quad \text{with } \Phi = 1 \tag{A5}$$

Consequently, for low Ni concentration we have $D^{\text{int}} = D_{\text{Ni}}$. Even if the interdiffusion coefficient would contribute to the scattering signal in the case of the $\text{Ge}^{60}_{98}\text{Ni}_2$ sample, this would only lead to an underestimation of the Ge self-diffusion coefficient (if we assume that Ni self-diffusion is slower than the Ge self-diffusion), and will not explain the observed difference between the diffusion coefficients determined on $\text{Ge}^{60}_{98}\text{Ni}_2$ and $\text{Ge}^{\text{nat}}_{98}\text{Ni}_2$ samples.

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