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Spontaneous generation of voltage in single-crystal Gd5Si2Ge2 during magnetostructural phase transformations

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Abstract

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Keywords

Materials Science and Engineering, gadolinium alloys, silicon alloys, germanium alloys, magnetomechanical effects, critical phenomena, solid-state phase transformations

Disciplines

Condensed Matter Physics | Metallurgy

Comments

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Spontaneous generation of voltage in single-crystal $Gd_5Si_2Ge_2$ during magnetostructural phase transformations

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The spontaneous generation of voltage (SGV) in single-crystal and polycrystalline $Gd_5Si_2Ge_2$ during the coupled magnetostructural transformation has been examined. Our experiments show reversible, measurable, and repeatable SGV responses of the materials to the temperature and magnetic field. The parameters of the response and the magnitude of the signal are anisotropic and rate dependent. The magnitude of the SGV signal and the critical temperatures and critical magnetic fields at which the SGV occurs vary with the rate of temperature and magnetic-field changes. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2162027]

Since their rediscovery in 1997,[1,2] the $R_5(Si_xGe_{1-x})_4$ intermetallic compounds, where R is a lanthanide metal, continue to attract considerable attention. This is because of their interesting physical properties, such as the giant magnetocaloric effect,[1–4] colossal magnetostriction,[5–8] giant magnetoresistance,[9–16] and spontaneous generation of voltage (SGV),[17] that have been observed during the first-order magnetostructural, ferromagnetic-orthorhombic to paramagnetic-monoclinic (on heating), phase transformation (see Refs. 5,18). Among them, the SGV is especially intriguing because it may result in the development of sensors, which can respond not only to changes in temperature, pressure, and/or magnetic field but most importantly to the rates of their changes without the need for a complicated analysis of signals. Furthermore, all of this can be done by a single sensor requiring no standby power.

The SGV in the $R_5(Si_xGe_{1-x})_4$ system was reported by Levin *et al.* in several polycrystalline samples of $Gd_5(Si_xGe_{1-x})_4$.[17] Because a current was not supplied, the voltages observed across the samples were regarded as spontaneous. The SGV occurs near first-order magnetostructural phase transformations. Here we report on the SGV during the first-order phase transition in several single-crystal $Gd_5Si_2Ge_2$ samples. In addition to the SGV behavior as a function of temperature and magnetic field, we also examine the anisotropy of the signal and its dependence on the variable rates of change of these stimuli.

The $Gd_5Si_2Ge_2$ single crystal was prepared using the Bridgman method.[19] Three parallelepiped-shaped samples were cut from a large grain by electric discharge machining. The longest side of each sample was parallel to one of the three major crystallographic directions [100], [010], or [001], and specimen dimensions were $3.66\times1.82\times0.49$, $4.06\times1.03\times1.00$, and $5.6\times2.0\times0.92$ mm^3, respectively. The SGV behaviors of a polycrystalline $Gd_5Si_2Ge_2$ sample with the dimensions of $6.57\times2.48\times1.85$ mm^3 were also measured in order to verify previous observations[17] and to compare them with the single-crystal samples.

The dc voltages across the samples were measured by a standard two-probe method and recorded as functions of time by a Keithley 181 nanovoltmeter. Readouts from the nanovoltmeter were computer recorded every 0.25 s. The temperature and magnetic-field changes exerted on the samples were regulated by a LakeShore Model 7225 magnetometer. The samples were subjected to the temperature and magnetic-field variations above and below their zero-field transition temperatures, at rates -7 to $+3$ K/min and -70 to $+70$ kOe/min. The magnetic fields were applied and the voltages were measured along the longest side of each sample. The misorientation between the directions of the magnetic-field vector and the crystal axes were less than ±5°. The SGV signal backgrounds caused by a minor thermal noise and drift of the nanovoltmeter were automatically subtracted before recording every sequence. The temperature readings from the sensor may deviate from the actual sample temperature by ±2 K due to the design of the magnetometer. The resolution of the Keithley 181 nanovoltmeter is 100 nV. The dc magnetization was measured using the same magnetometer.

The SGV signals of a $Gd_5Si_2Ge_2$ single-crystal sample along the [010] direction as functions of temperature and the rate of temperature change in a zero magnetic field are displayed in Fig. 1. The characteristics of the temperature-induced SGV signals are similar for all three samples. The onset temperatures of the SGV signals are located within 2 K of the Curie temperature (T_C) which are 269 and 259 K for warming and cooling, respectively, confirming that the origin

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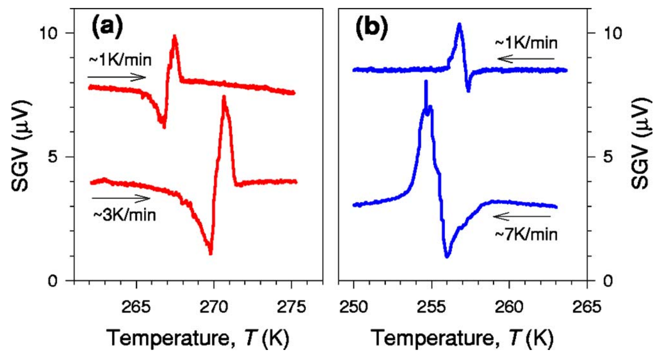

FIG. 1. (Color online) The SGV as a function of temperature for a $Gd_5Si_2Ge_2$ single-crystal sample measured along the [010] direction upon heating at ~1 and ~3 K/min (a) and upon cooling at ~1 and ~7 K/min (b) in a zero magnetic field.

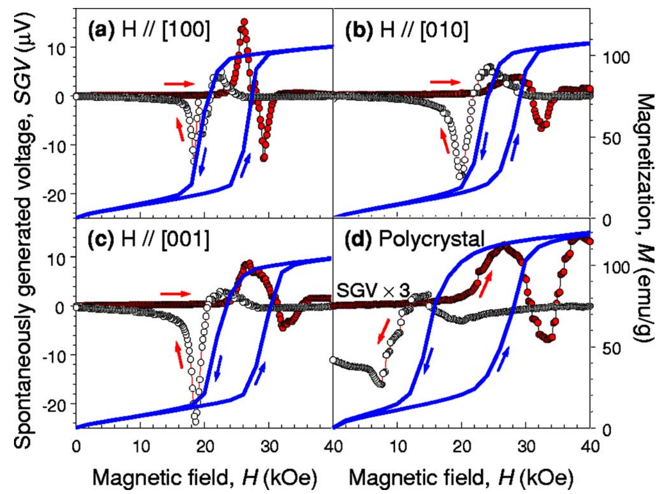

FIG. 2. (Color online) The SGV and dc magnetization as functions of a magnetic field measured for $Gd_5Si_2Ge_2$ along the [100] (a), [010] (b), and [001] (c) directions and for a polycrystal (d). The solid and open circles represent the SGV signals upon increasing field and decreasing field, respectively. The lines without symbols represent the dc magnetization. All measurements were carried out at ramping rates of +40 and −40 kOe/min when samples were held ~9 and ~2 K above their T_C's for the single crystals and polycrystal, respectively. The T_C's were determined from the dc magnetization measured upon warming in a 1 kOe field, which are 269, 269, 275, and 276 K for the [100], [010], and [001] directions and the polycrystal, respectively.

of the SGV is the coupled magnetic and crystallographic phase transformation. A thermal hysteresis of about 10 K of the SGV signals is observed. All signals are S shaped, starting at T_C and ending at temperatures 2–4 K higher or lower than T_C upon heating or cooling, respectively. A higher-temperature sweep rate increases the magnitude of the SGV signal and raises or lowers the temperature at which the SGV signal appears upon heating or cooling, respectively. The shapes of the SGV signals are quite similar to the thermal emf signals observed during the freezing of water, the melting of tin, and the solidification and the two polymorphic transformations of CuBr, suggesting that a similar mechanism may be responsible for these signals.[20]

Figure 2 shows the isothermal SGV signals induced by sweeping the magnetic field at a rate of ~40 kOe/min in the single-crystal samples along three principal crystallographic directions and in a polycrystalline sample. The isothermal dc magnetization data measured at the same temperatures are also shown in Fig. 2. Because there are compositional differences in different single-crystal samples (see Ref. 16 for details), the temperatures were normalized to the individual T_C's determined for each specimen from low-field dc magnetization. The onsets and offsets of the SGV signals triggered by magnetic fields coincide, respectively, with the rapid increase of the magnetization and its saturation due to the magnetic-field-induced phase transitions between the paramagnetic and ferromagnetic states. Upon field increasing, the SGV signal starts at the onset of the paramagnetic-monoclinic (PM) to ferromagnetic-orthorhombic (FM) transition, reaches a maximum and then drops to a minimum, and finally goes back to zero when the magnetic transition is complete. Upon field decreasing, the shape of the SGV signals does not change much, with a maximum appearing first, followed by a minimum. The onset and end points of the SGV signals occur at lower fields during demagnetizing than during magnetizing, which is normal for a first-order phase transformation. The magnetic fields of the onsets and especially of the ends of the SGV signals are slightly different for different crystallographic directions, demonstrating weak anisotropy.

The shapes of the SGV signals are similar for the [010] and [001] direction samples when subjected to a same variation of a same trigger (temperature or magnetic field), while that of the [100] direction sample seems unique. As it is known from the structural characterization of the $Gd_5Si_2Ge_2$, the shear movement of the slabs during the coupled magnetostructural phase transformation occurs along the [100] direction,[18] and this is likely the reason for the uniqueness of the SGV behavior of the [100] direction sample. There is a small difference in the magnitudes of the SGV signals for the single-crystal samples along different crystallographic directions. Since the magnitude of the SGV signal depends on the sample shape and the distance between the two electrical connections to the sample, we believe that this small difference in the magnitudes is extrinsic. The magnitudes of the SGV signals for the polycrystalline sample, however, are about three times smaller than those of the single-crystal samples despite similar shapes and locations of the electrical connections to the samples. Thus, a smaller SGV signal in polycrystal is intrinsic. This feature correlates well with the fact that the first-order transitions in single-crystal samples are usually sharper than those in polycrystalline samples of the same composition, and it is in line with the results from the magnetic force microscopy and thermal expansion studies of a $Gd_5Si_2Ge_2$ single crystal.[21]

As mentioned above, the higher-temperature sweep rate increases the magnitude of the SGV signal. For the magnetic-field-induced SGV signals, the increase of the rate of change of the stimulus also increases the magnitude of the signal, as shown in Fig. 3(a). The critical field at which the SGV signal starts also exhibits a systematic change with changing the magnetic-field sweep rate. As shown in Fig. 3(b), the increased field sweep rate shifts the critical field to lower values for all the samples. This is in line with an earlier observation that the critical field of the magnetostructural transition in Gd_5Ge_4 is also field sweep rate

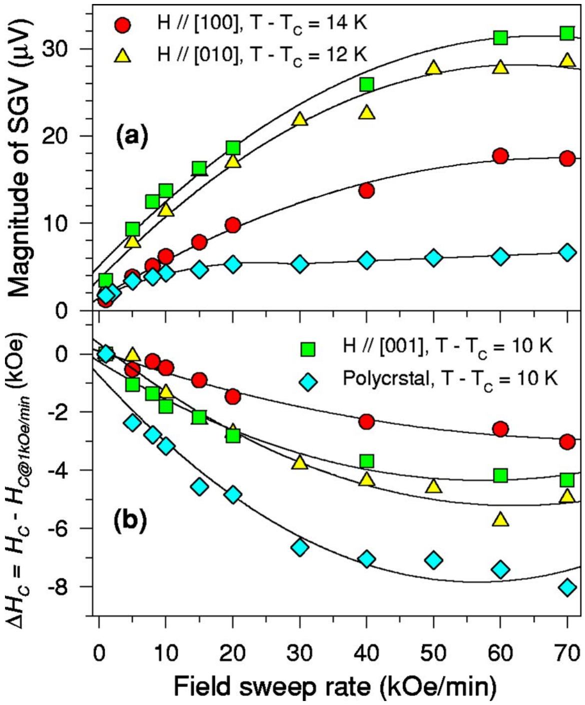

FIG. 3. (Color online) The magnitude of the SGV (a) and the change of the critical magnetic field of SGV relative to the critical field at 1 kOe/min (b) as functions of the magnetic-field sweep rate in single-crystal and polycrystalline $Gd_5Si_2Ge_2$ samples measured isothermally at temperatures from ~10 to ~14 K above their T_C's, which were measured upon warming in a 1 kOe field. The lines are guides to the eye.

dependent.[22] There are some differences in the absolute values of the magnitudes of the signals and the values of the $\Delta H_C = H_C - H'_C$, where H'_C is the critical magnetic field at 1 kOe/min for different samples, but we believe that they may be extrinsic for different single-crystal samples. The difference between the polycrystalline and single-crystal samples is intrinsic due to the reasons mentioned above.

The main reason for the SGV was thought to be the Seebeck effect, given the similarity of the SGV profile and the signal recorded from a differential thermocouple simultaneously.[17] A compositional difference or/and a temperature difference on the two ends of the sample triggers the phase transformation from one specific end, propagating to another. The heat release or absorption during this process gives rise to the S shape and polarity change of the SGV signal when the direction of temperature or/and magnetic-field change is reversed. It is worth noting, however, that when a sample is flipped without reversing the polarity of the electrical connections, the polarity of the SGV signal also reverses. This behavior supports the notion that there is a small gradient of composition along the sample, and therefore a slight change of the T_C, because a temperature gradient caused by the instrument should always be the same, regardless of how the sample is mounted.

In summary, the spontaneous generation of voltage in both single-crystal and polycrystalline $Gd_5Si_2Ge_2$ samples has been studied as a function of temperature, magnetic field, and rates of their changes. The critical temperatures and magnetic fields at which the SGV signals appear nearly coincide with the Curie temperatures and the critical magnetic fields at which the first-order magnetostructural phase transformations occur. This observation confirms the intimate relationship between the SGV and the coupled magnetic crystallographic phase transformation. A weak anisotropy was observed in single-crystal $Gd_5Si_2Ge_2$ samples through their different SGV responses. The magnitude of the SGV signal clearly increases with the rate of the temperature and magnetic-field change. The rate of stimuli changes also shifts the critical temperatures and critical magnetic fields. Although more research is needed to better map out the details of the SGV behavior in the $Gd_5Si_2Ge_2$ alloy, it is a likely candidate for a smart temperature and magnetic-field sensor material for near room-temperature applications, adding another aspect of its potential manifold applications.

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