Spontaneous generation of voltage in single-crystal Gd$_5$Si$_2$Ge$_2$ during magnetostructural phase transformations

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Abstract
The spontaneous generation of voltage (SGV) in single-crystal and polycrystalline Gd$_5$Si$_2$Ge$_2$ 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.

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$_5$Si$_2$Ge$_2$ during magnetostructural phase transformations

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The spontaneous generation of voltage (SGV) in single-crystal and polycrystalline Gd$_5$Si$_2$Ge$_2$ 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.

Since their rediscovery in 1997, the R$_5$(Si$_{1-x}$Ge$_x$)$_4$ 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, colossal magnetostriction, giant magnetoresistance, and spontaneous generation of voltage (SGV), that have been observed during the first-order magnetostructural transformation in several single-crystal and polycrystalline Gd$_5$Si$_2$Ge$_2$ samples. In addition to the SGV behavior as a first-order phase transition in several single-crystal Gd$_5$Si$_2$Ge$_2$ samples, the SGV signals of a Gd$_5$Si$_2$Ge$_2$ 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...
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.

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 from the 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 demagnetization 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$_5$Si$_2$Ge$_2$, the shear movement of the slabs during the coupled magnetostructural phase transformation occurs along the [100] direction, 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$_5$Si$_2$Ge$_2$ single crystal.

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$_5$Ge$_4$ is also field sweep rate
magnetic fields at which the SGV signals appear nearly co-
and rates of their changes. The critical temperatures and
has been studied as a function of temperature, magnetic field,
compositional difference or/and a tem-
Seebeck effect, given the similarity of the SGV profile and
difference between the polycrystalline and single-crystal
1 kOe/min for different samples, but we believe that they
fore a slight change of the
electrical connections, the polarity of the SGV signal also
gives rise to the S shape and polarity change of the SGV
other aspect of its potential manifold applications.
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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 polycrys-
talline Gd₅Si₂Ge₂ samples measured isothermally at temperatures from −10
to −14 K above their Tₛ/𝑠, which were measured upon warming in a 1 kOe
field. The lines are guides to the eye.

There are some differences in the absolute values of the magnitudes of the signals and the values of the
ΔHₛ = Hₛ − Hₛ', where Hₛ' 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 dif-
ference 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. A compositional difference or/and a tem-
tperature difference on the two ends of the sample triggers the
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 there-
therefore a slight change of the Tₛ because a temperature grad-
ient 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 Gd₅Si₂Ge₂ 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 co-

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