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ABSTRACT

Gallium oxide has recently been identified as a promising scintillator. To assess its potential as a detector material for ionizing radiation at low temperatures, we measured the luminescence and scintillation properties of an undoped Ga_2O_3 crystal over the 7–295 K temperature range. The emission of the crystal is due to the radiative decay of self-trapped excitons and donor-acceptor pairs and peaks at a wavelength of 380 nm. The scintillation light output of the undoped Ga_2O_3 increases with a decrease in temperature, reaching a maximum value of 19 300 ± 2200 ph/MeV at 50 K. The measured luminescence kinetics has a recombination character with specific decay time ($\tau_{0.1}$) increasing from 1 to 1.8 μs at cooling. Since radiative decay in the crystal competes with nonradiative processes, material optimization could lead to the scintillator achieving a yield of 40800 ph/MeV, a figure considered to be an upper limit.

X-rays, γ-quanta, and particles (electrons, neutrons, alpha) are extensively used for scientific explorations and in many applications related to medicine, industry, security, and environmental monitoring. The scintillation method, involving the conversion of deposited energy to light, is widely used for the detection of high-energy ionizing radiation. The critical advantage of a scintillation detector is its nonreliance on the extraction of charged particles from the material. Scintillation light can be detected directly from the bulk of the crystal absorber. Here, it is generated by radiative decay of excited particles with a very fast response, characteristic of exciton emission. Therefore, the development of more efficient materials for scintillation detection remains an important topic in relevant areas of modern scientific research. The topical review[1] gives a good overview on the subject.

Semiconductor materials are of particular interest as scintillators due to their generally high conversion efficiency which translates directly into enhanced energy resolution. This is caused by the lower bandgap compared with that typical for dielectric scintillators and the higher mobility of free carriers. The light yield per unit of absorbed energy is inversely proportional to the value of the energy gap of a material,[2,3] so that the decrease in this parameter results in higher scintillation efficiency. Moreover, the high mobility of free electrons and holes in semiconductors ensures their prompt escape from the region of the ionization track, with high probability of nonradiative quenching.[4,5] This facilitates the transfer of excitation energy to the activators. Consequently, doped semiconductor scintillators, such as ZnS-Ag,[6] ZnSe-Te,[7] and ZnTe-O,[8] demonstrate high light yield at room temperature, warranting their application in detectors for high-energy photons and particles. Another example is Ga-doped ZnO, exhibiting high light yield and subnanosecond decay time constant due to exciton emission.[9] Recently, the scintillation properties of undoped Ga_2O_3 have been examined for the first time, and it was shown that the material is a promising scintillator, exhibiting a light yield of 15 000 ph/MeV and a decay time constant of 8 ns at room temperature.[10] This triggered further investigations of the material scintillation properties.[11,12]

This finding also prompted us to investigate the performance of this material as cryogenic scintillator, driven by the development of techniques for particle detection at low temperature. The latter is currently motivated by the requirements of experiments in fundamental physics searching for neutrinoless double beta decay[13] and dark matter particles.[14] Aiming to achieve maximum sensitivity, some of these experiments use cryogenic phonon-scintillation detectors that feature both high-energy resolution and event discrimination capability.[15–17] The principal advantage of this technique lies in a significant (by few

orders of magnitude) reduction of contributions from radioactive background. Recent achievement is demonstrated by the latest results from cryogenic experiments searching for neutrinoless double beta decay[18] and dark matter particles.[19] There is continuing interest in furthering this technique and finding new scintillation materials suitable for low temperature application and is an important aspect of related research.[3,15,20] Furthermore, this activity creates a foundation for the exploration of new concepts and applications, of which the most appealing is harnessing the time-of-flight detection method for positron emission tomography using fast and bright cryogenic scintillators.[21,22]

In this work, we carried out measurements of X-ray luminescence, decay kinetics, and scintillation light output of undoped Ga_2O_3 as a function of temperature. By exploring and analyzing the temperature dependence of these properties, we extended our knowledge of the material and assessed the prospect of Ga_2O_3 for application as cryogenic scintillation detector. These studies provide additional valuable information about the processes that transform absorbed energy into scintillation, which also improves the general understanding of the scintillation mechanism in solids at fundamental levels.

The sample of undoped β-Ga_2O_3, used in this study, was obtained from Tamura Co. (Japan). For the luminescence measurements, the sample was placed into a closed-cycle He cryostat, equipped with a DE-202A cryocooler (Advanced Research Systems) and Cryocon 32 (Cryogenic Control Systems Inc.) temperature regulator. The steady-state X-ray luminescence was excited by a URS-55A X-ray tube with a Cu-anticathode tube operating at 55 kV and 10 mA. The emission spectra were recorded using an MDR-12 spectrograph with a spectral resolution of 1 nm and a Hamamatsu H9305 photomultiplier module, sensitive over a 200–700 nm wavelength range.

For measurements of scintillation characteristics, the crystal was placed in a helium constant flow cryostat and excited by α-particles from an ^{241}Am source. The measurements and analyses were carried out using the multiphoton counting technique.[23] The signal, detected by a multialkali photomultiplier model 9124 A (Electron Tubes Enterprises, Ruislip, UK), was digitized by a fast analogue-to-digital converter (ADC) with a 5 ns sampling interval. This allows resolving individual photons and recording single photon signals (SPSs). The technique enables studies of scintillation processes with decay time constants in the range of 10^{-6}–10^{-3} s. Spanning such a wide range when changing the operating temperature is quite common in many scintillation materials. Furthermore, it allows measuring both decay time and light yield characteristics of scintillators in a single experiment. It is therefore well suited for the investigation of temperature-dependent scintillator properties as documented in various publications.[3,23,24]

The most common β-polymorph of gallium oxide has a monoclinic crystal structure (space group C2/m) with lattice constants a = 12.214 Å, b = 3.037 Å, c = 5.798 Å, $\alpha = 90°$, $\beta = 103.83°$, $\gamma = 90°$.[25] The crystal unit cell contains GaO_6 octahedral and GaO_4 tetrahedral chains aligned along the b-axis. Oxygen atoms are located in three crystallographically different positions and form a distorted cubic closed packed array. It is a wideband semiconductor with a bandgap energy $E_g = 4.85$ eV, a density of 5.95 g/cm^3, and a melting point of 1795 °C.[26]

Under steady-state X-ray excitation, Ga_2O_3 exhibits a broad emission band in the UV-green region of emission spectrum that gradually increases in intensity with cooling the crystal (see Fig. 1).

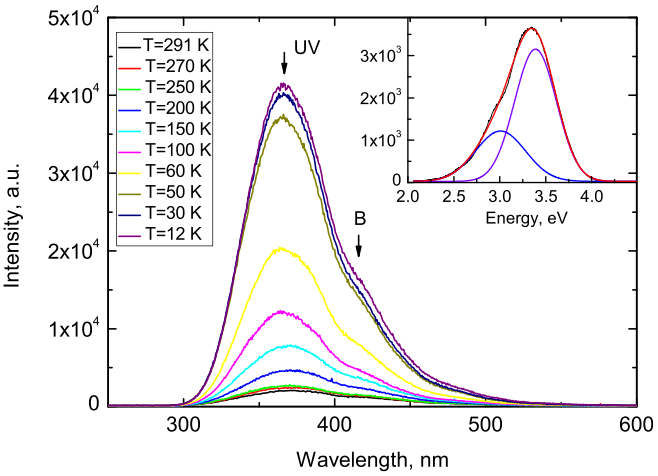

FIG. 1. X-ray luminescence spectra of Ga_2O_3 measured at different temperatures. Two emission bands are marked as UV and B. The inset shows the emission spectrum at T = 12 K as a function of energy, decomposed into two Gaussians peaking at 3.01 eV (blue emission) and 3.38 eV (UV emission), respectively.

The emission band peaking at 380 nm has a complex character, which is a characteristic for a system where transitions occur within different emission centers. This is demonstrated in the inset of Figure 1, where the emission spectrum measured at a temperature of 12 K is plotted as a function of photon energy.

The spectrum can be represented as a sum of two Gaussians at 3.01 and 3.38 eV, indicating that at least two types of emission centers exist in the crystal under study. Such a composite structure of luminescence spectra is a characteristic feature of gallium oxide. Previous studies of luminescence properties of undoped Ga_2O_3 reported at least three strongly overlapping emission bands in UV, blue, and green parts of the emission spectra.[27–30] The UV band is generally independent of specific impurities and thus is assigned to the emission of self-trapped excitons. This emission typically manifests fast kinetics in the nanosecond range. The blue and green bands are attributed to transitions involving deep donors and acceptors. Electrons recombining with holes at the defect sites are the origin of recombination luminescence with long decay kinetics (microsecond range). The formation energy of different types of intrinsic defects in β-Ga_2O_3 was analyzed in a recent theoretical work.[31] The results of that study confirmed that blue emission is due to complexes of oxygen and gallium vacancies (V_O+V_{Ga}).

Further, we studied the scintillation properties of undoped Ga_2O_3 over the temperature range of 7–295 K. The crystal exhibits a good scintillation response allowing to detect a peak due to α-particles from an ^{241}Am source depositing their kinetic energy in the scintillator. The resulting pulse height spectra of the crystal at different temperatures are displayed in Fig. 2. Given that scintillation light yield is proportional to the position of this peak, the variation of the light output of the crystal under investigation with temperature can be traced by monitoring the change in the peak position. The temperature dependence of the light output for the undoped Ga_2O_3 derived in this way is shown in Fig. 3. With cooling the crystal, the peak shifts toward higher amplitudes, indicating a steady rise of the scintillation light output. This trend is observed until the temperature is lowered to 50 K,

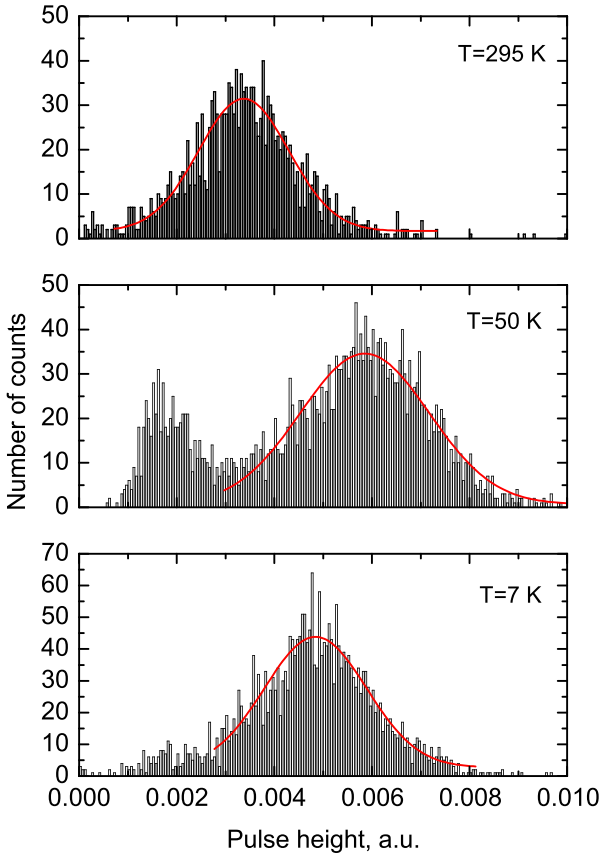

FIG. 2. Pulse height spectra of Ga_2O_3 at T = 7, 50, and 295 K, measured at the excitation with α-particles from ^{241}Am. The curve lines show the Gaussian fitting the spectra. The feature observed at the low-energy side of the α-peak at low temperatures is due to the gradual cut-off of background by a trigger affected by the baseline nose.

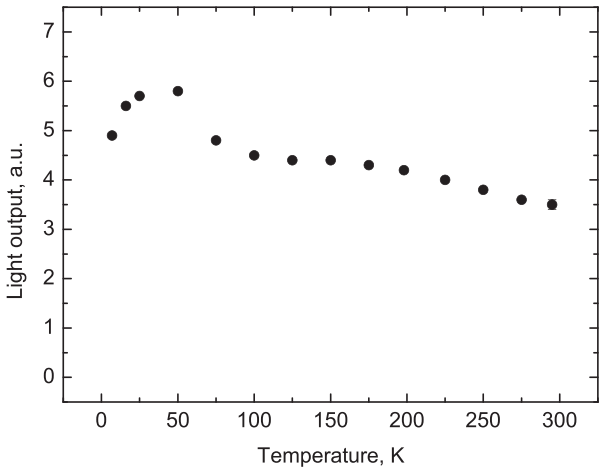

FIG. 3. Scintillation light output of Ga_2O_3 as a function of temperature, measured for the excitation with α-particles from ^{241}Am.

below which the scintillation light output starts to decrease, as shown in Fig. 3. At 7 K, the measured scintillation light output of the Ga_2O_3 crystal reduces to about 80% of its maximum value. It is worthwhile noting that the decrease in scintillation light yield at very low temperature has been observed in some other scintillators. One explanation of this feature is a capture of excited carriers by shallow traps[32,33] that cause long-lasting afterglow. Due to charge trapping, the excitation energy is not anymore released during the actual scintillation event but over a much longer period of time (10^{-3}–1 s) as phosphorescence signal and the corresponding fraction of excitation cannot therefore be detected, thus resulting in a decrease in the scintillation output.

Figure 4 shows the scintillation decay curves of undoped Ga_2O_3 monitored at different temperatures. We measured scintillation decay curves in the integral regime, capturing the entire emission spectrum of the scintillator, but due to the limited timing resolution of the measuring technique (0.1 μs), the nanosecond component reported by Yanagida *et al.*[10] cannot be identified. The existence of several types of emission centers with different types of decay kinetics results in a complex nonexponential decay that is a characteristic feature of recombination luminescence.[34] This complexity of the scintillation process imposes certain limitations on the analysis of the decay kinetics. More specifically, the fitting of the scintillation decay curves using a linear combination of exponential functions is merely a mathematical way of representing the experimental results. In such a case, it is not possible to relate the fitting parameters directly to the physical quantities that describe the specific emission processes. Therefore, in such a case, more practical quantitative characteristic of the decay is a time when the initial intensity decreases by an order of magnitude ($\tau_{0.1}$). The temperature variation of the $\tau_{0.1}$ in the crystals under investigation is shown in the inset of Fig. 4. The shape of the scintillation curves is largely unaffected by temperature; the only noticeable change—the emergence of a long component—is observed at T < 60 K. We attribute this component to the abovementioned process of recombination of carriers released from the shallow traps. Otherwise, this behavior is consistent with what has been seen in other semiconductor scintillators.[6,35]

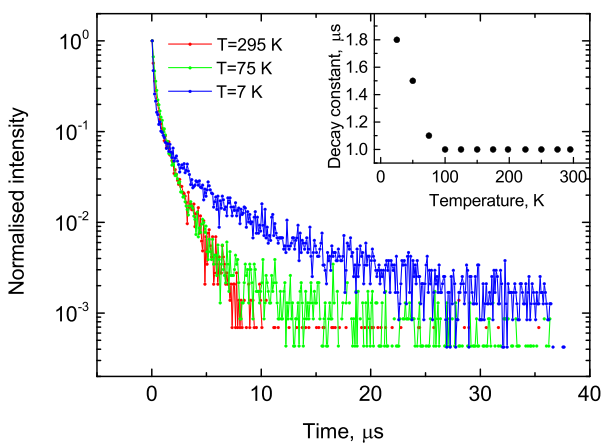

FIG. 4. Normalized scintillation decay curves of Ga_2O_3 at T = 7, 75, and 295 K measured at the excitation with α-particles from ^{241}Am. The inset shows the temperature dependence of the decay constant $\tau_{0.1}$.

To assess the performance of the Ga_2O_3 as the scintillator, it is essential to quantify the light yield of the material. First, we calculated the theoretical limit for the absolute light yield of the Ga_2O_3 scintillator using the semiempirical approach developed by Lempincki *et al.*[2] and later refined in Ref. 3. In this case, the energy transfer efficiency and luminescence quantum efficiency are assumed to be equal to 1, bringing about the following equation for the absolute light yield of a scintillator:

$$LY = \frac{10^6}{2.35E_g}\left[1 + 0.158 \times 10^4\left\{\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0}\right\}\frac{(h\nu_{LO})^{\frac{3}{2}}}{1.5E_g}\right]^{-1} (\text{ph/MeV}). \quad (1)$$

Here, $\varepsilon_0 = 11.2$ and $\varepsilon_\infty = 3.8$[26] are the static and high-frequency relative permittivities, respectively, of the material, and $h\nu_{LO} = 99$ meV[36] is the maximum energy of longitudinal optical (LO) phonons. Substituting the numerical values into Eq. (1), we obtained an upper limit for the light yield of Ga_2O_3 equal to 40 800 ph/MeV.

We also measured the light yield of Ga_2O_3 experimentally. Typically, the light yield is derived by comparing the scintillation response of the crystal under study with that of a reference scintillator measured under identical experimental conditions. This approach assumes that the light collection efficiency of the experimental setup used for the measurements is a constant parameter determined by geometrical factors that are identical. In this case, we used as a reference an $Lu_{2(x-1)}Y_{2x}SiO_4$-Ce (LYSO-Ce) scintillator, known for its high light yield (34 000 ph/MeV) and fast decay time (33 ns), both changing only insignificantly with cooling.[37,38] This scintillator emits in the same spectral range as Ga_2O_3, which is beneficial for reducing the errors arising from the uncertainty of calculating the emission-weighted detector efficiency ε_λ.[39,40] This parameter accounts for the difference in the spectral sensitivity of the detector and was calculated from the measured X-ray luminescence spectra of the crystals and the known quantum efficiency of the 9124A photomultiplier.

The value of ε_λ was found to be 0.27 and 0.25 for Ga_2O_3 and LYSO-Ce, respectively. Under the aforementioned assumption of identical light collection efficiency, the light output measured in the experiment is proportional to the two variables, i.e., absolute light yield (LY) and emission-weighted detector sensitivity:[39,40] $N_m \sim LY \times \varepsilon_\lambda$. Thus, from the measurements obtained with the two scintillators, we can estimate the light yield of Ga_2O_3 using

$$LY_{Ga2O3} = LY_{LYSO} \times \frac{N_{Ga2O3}}{N_{LYSO}} \times \frac{\varepsilon_{\lambda,\,LYSO}}{\varepsilon_{\lambda,\,Ga2O3}}. \quad (2)$$

Taking into account a typical value of the absolute light yield of LYSO-Ce, we determined that the light yield of Ga_2O_3 is equal to 11 600 ± 1600 ph/MeV at 295 K, reaching a maximum value of 19 300 ± 2200 ph/MeV at 50 K. The error of this evaluation comes mainly from the uncertainty in the position of the peak that corresponds to α-events in the measured scintillators. This finding evidences that undoped β-Ga_2O_3 can be used for scintillation detection over a wide temperature range. Comparison of the measured light yield with the theoretical prediction also shows that this parameter for the measured crystal is only half of what it can be, and hence gallium oxide has a significant potential for further improvement of its scintillation properties.

In this work, we report the scintillation and luminescence properties of β-Ga_2O_3 measured as a function of temperature down to 7 K. Two emission bands observed in the UV and blue spectral ranges are assigned to the radiative recombination of self-trapped excitons and donor-acceptor pairs, respectively. The scintillation light output of the crystal increases progressively with a decrease in temperature, reaching a maximum value of 19 300 ± 2200 ph/MeV at 50 K, from where it reduces by 30% at further cooling to 7 K. It is found that at room temperature, undoped Ga_2O_3 exhibits about one-third of the light output of a commercial LYSO-Ce scintillator. The detection efficiency of the crystal is lower in comparison with the champions in the field, but taking into consideration the fact that intrinsic defects play a major role in the emission of gallium oxide, we envisage that the substantial improvement of the scintillation properties is likely to occur through material doping and optimization of production technology.

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