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Calculation of electric field gradients and site preference in Ba-Al-Ge clathrates SERGIO RODRIGUEZ, WEIPING GOU, JOSEPH ROSS, Texas A&M University — Sn, Ge or Si can form cage-like clathrate structures, many of which exhibit enhanced thermoelectric performance. To understand Al substitutional occupation in $Ba_8Ge_{46-x-y}Al_x\square_y$ clathrates we performed NMR lineshape simulations for $3 \leq x \leq 24$, and $0 \leq y \leq 3$, where $\square$ represents a vacancy. The electric field gradient (EFG) was calculated for Al sites assuming an ordered superstructure. To obtain the EFG we used *ab initio* methods in the Generalized Gradient Approximation as implemented by the WIEN2k code with structural relaxation. Results where used to simulate NMR lineshapes numerically. These were compared to our previously reported NMR lineshapes. For fully occupied $Ba_8Ge_{30}Al_{16}$ we compare different site occupations, obtaining good agreement and thus information about Al site preferences. For reduced-Al samples, WDS measurements indicate the presence of spontaneous vacancies. In the case of the Zintl phase $Ba_8Ge_{33}Al_{12}\square_1$ we found that Al sites adjacent to the vacancy exhibit a large EFG, while those with the vacancy further away have smaller EFG's. Assuming a larger Knight shift for sites next to vacancies, we obtain good agreement with NMR experimental results for reduced-Al $Ba_8Ge_{34}Al_{12}$. We infer that Al prefers locations close to vacancies rather than random occupation. Supported by Robert A. Welch Foundation (Grant A-1526).

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