

# Nuclear Density Analysis by Using Sparse Modeling

HIROSHI TANAKA

DEPARTMENT OF PHYSICS AND MATERIALS SCIENCE, SHIMANE UNIVERSITY

## ABSTRACT

Recently, the author of this article and his colleagues have succeeded in developing a novel method for evaluating nuclear density in detail from neutron diffraction data by employing sparse modeling (SM). The method was applied to  $\text{KH}_2\text{PO}_4$  and subsequently it was shown that the hydrogen distribution in  $\text{KH}_2\text{PO}_4$  has double maxima; this is a result that has been observed only faintly by conventional analyses such as the maximum entropy method (MEM). Furthermore, the computational time of SM is comparable to or shorter than that of the MEM. Thus, SM has the potential to replace the MEM as a method for evaluating nuclear density.

## INTRODUCTION

Up until recently, a scheme using the maximum entropy method (MEM) [1] has been used frequently in order to evaluate the nuclear density from neutron diffraction data, and this scheme has produced many achievements. The MEM tends to choose a nuclear density that varies the most gradually in space among nuclear densities that reproduce the diffraction data obtained by experiment. The nuclear density is, however, quite localized in many cases and therefore the MEM seems to be inappropriate for evaluating nuclear density in those cases.

Consequently, we employed sparse modeling (SM) as an alternative to the MEM. SM assumes sparsity of data, and tends to choose sparse (i.e., localized) ones among those reproducing the results observed by experiment.

## METHOD

SM originally developed in the field of signal processing, and has been applied in situations such as image restoration and the reconstruction of tomographic images by MRI. The application of SM to the analysis of radio telescope data is fresh in our memory, and resulted in the visualization of a black hole. In a mathematical sense, it is a method to find out the optimal solution in the multivariate linear optimization problem, assuming that values of most variables are zero (sparsity) [2].

First, we define the  $\ell^p$  norm of discretized nuclear densities  $\{\rho_i\}$  in real space by

$$\|\rho\|_p = \left( \sum_i |\rho_i|^p \right)^{1/p}$$

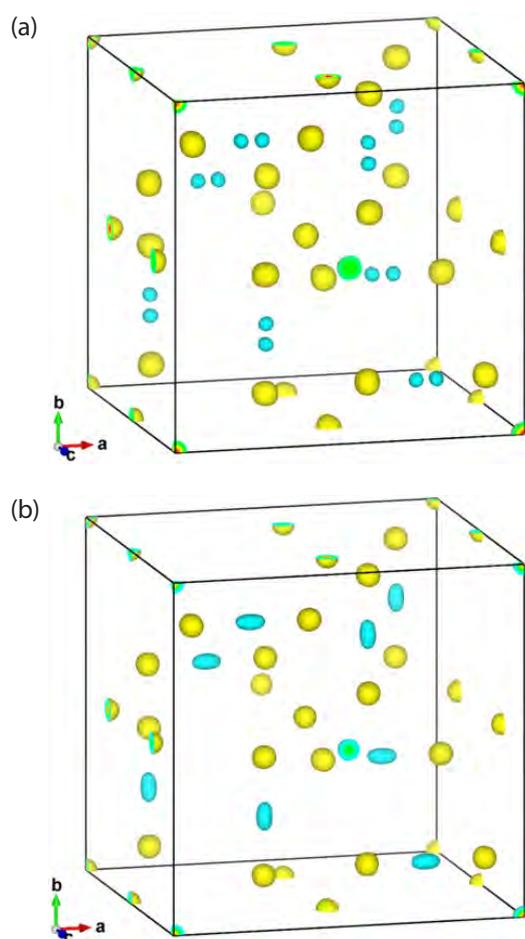
In SM, the scheme called LASSO (least absolute shrinkage and selection operator) [3] is often used, in which sparsity is achieved by minimizing the  $\ell^1$  norm  $\|\rho\|_1$ . However, we cannot use this scheme because the nuclear density  $\{\rho_i\}$  satisfies a sum rule and its  $\ell^1$  norm is constant. It is known that a sparse solution will be obtained by choosing a value of  $p$  within the range  $0 \leq p \leq 1$ . We, therefore, formulated a new scheme of SM using the  $\ell^{1/2}$  norm and obtained a proximal operator for the  $\ell^{1/2}$  norm, which is needed for an iterative calculation to achieve the sparse solution.

## APPLICATIONS

We applied our method to  $\text{KH}_2\text{PO}_4$  (KDP), a typical ferroelectric material, in the paraelectric phase. It is known

that hydrogen distribution shows symmetric double maxima between two oxygen atoms at room temperature. One of the double maxima, however, disappears as temperature decreases, and the hydrogen distribution becomes asymmetric. Along with this, KDP undergoes a phase transition and becomes ferroelectric. We used neutron diffraction data in Ref. [5], which were taken at JRR-3M for non-deuterated KDP in powder at room temperature.

Figure 1 (a) shows the isosurfaces of the nuclear density at  $20 \text{ \AA}^{-3}$  (colored in yellow) and at  $-20 \text{ \AA}^{-3}$  (colored in light blue) analyzed by SM. The isosurfaces are compared to the results of the MEM (b) using ENIGMA [6]. The isosurfaces were drawn by the visualization program VESTA [7]. As a whole, they show good agreement with each other except for the area around the hydrogen atoms, colored in light blue.



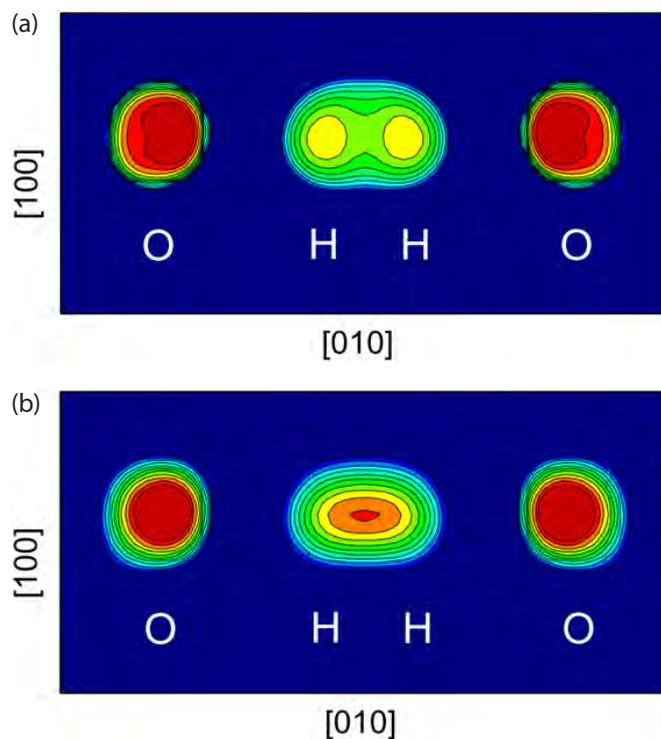
**Fig. 1:** Comparison of the isosurfaces of nuclear density for  $\text{KH}_2\text{PO}_4$  at  $20 \text{ \AA}^{-3}$  (colored in yellow) and at  $-20 \text{ \AA}^{-3}$  (colored in light blue) obtained by SM (a) and by the MEM (b).

In order to investigate the differences in nuclear density around hydrogen atoms, we compared the contour map of the nuclear density analyzed by SM (a) with that of a map analyzed by the MEM (b), as seen in Fig. 2 on the plane where two oxygen atoms and two hydrogen atoms are aligned in a straight line. It is clearly shown that hydrogen distribution shows double maxima in the result that uses SM. On the other hand, the double maxima distribution is blurred out and difficult to recognize in the result that uses the MEM. It is also observed that the nuclear density around oxygen atoms is asymmetric due to hydrogen bonding, which is not so clear in the result using the MEM.

We also compared the computational time of SM with that of the MEM. Although it depends on several conditions, the computational time of SM is comparable to or shorter than that of MEM by three to five times. This means that SM is also superior to the MEM in analysis speed.

## SUMMARY

We developed a new method evaluating the nuclear density from neutron diffraction data by using SM. SM



**Fig. 2:** Comparison of the contour maps for  $\text{KH}_2\text{PO}_4$  on the plane where 2 oxygen atoms and 2 hydrogen atoms are aligned in a straight line, using analyses obtained by SM (a) and by the MEM (b). The contour lines are drawn at  $2^{n/2} \text{ \AA}^{-3}$  ( $n = 2, 3, \dots, 12$ ).

can reproduce nuclear densities that are more precise or probable than the MEM because the nuclear density is sparse in general.

After several additional tests, the program we developed will be released for free. We hope it is useful for the analysis of neutron diffraction.

**Acknowledgements:** The author would like to thank Prof. Masaki Takata of Tohoku University for providing the neutron diffraction data of  $\text{KH}_2\text{PO}_4$  (KDP).

### References

- [1] M. Sakata, T. Uno, M. Takata, and C.J. Howard, *J. Appl. Crystallogr.* 26, 159 (1993).
- [2] A.M. Bruckstein, D.L. Donoho, and M. Elad, *SIAM Rev.* 51, 34 (2008).
- [3] R. Tibshirani, *J. R. Statist. Soc. B* 58, 267 (1996).
- [4] H. Tanaka, M. Oie, and K. Oko, *J. Phys. Soc. Jpn.* 88, 053501 (2019).
- [5] S. Kumazawa, S. Yamamura, E. Nishibori, M. Takata, F. Izumi, and Y. Ishii, *J. Phys. Chem. Solids* 60, 1407 (1999).
- [6] H. Tanaka, M. Takata, E. Nishibori, K. Kato, T. Iishi, and M. Sakata, *J. Appl. Crystallogr.* 35, 282 (2002).
- [7] K. Momma and F. Izumi, *J. Appl. Crystallogr.* 44, 1272 (2011).

Reference 4 was published in the April 2019 issue of the *Journal of the Physical Society of Japan* as an Editor's Choice article.



**Hiroshi Tanaka** is a professor at the Department of Physics and Materials Science, Shimane University. He received a PhD from the University of Tokyo while working at IBM Tokyo Research Laboratory, and subsequently became a professor at Shimane University in 1996. His research interests are in the theory of condensed matter physics, mathematical physics and computational physics.