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SIZE DISTRIBUTION AND SURFACE EFFECTS IN ULTRAFINE PARTICLE NGR SPECTRA

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Résumé.- Les spectres Mössbauer de goethite en petites particules sont comparés aux prévisions de deux modèles : (1) le modèle de Mørup qui corrèle distribution de champ et distribution de taille des particules, (2) un modèle où la réduction du champ est attribuée à des effets de surface. Le second modèle semble mieux expliquer l'influence de la température.

Abstract.- Presently two different approximations exist in order to explain the asymmetrically broadened lineshapes observed in ultrafine iron oxides and iron oxy-hydroxides. One, due to Mørup et al. /1/ uses a simple pre-relaxation model for the total sublattice spin of the particle, together with a particle size distribution. By adjusting suitable distribution parameters and the anisotropy constant, indeed a reasonable representation of the experimental results can be obtained. The second approach, as we proposed in previous papers /2,3/ considers the observed reduced magnetic fields mainly to be due to the surface cations being gradually decoupled from the interior of the particle. Unfortunately neither a detailed shape of this field reduction near the surface can as yet been given, nor a clear description of the changes of the Mössbauer fraction f near the surface. However here too a field function can be proposed and suitable parameters be adjusted to match the experimental results; these parameters are the maximum inside field H_i^0 , a Gaussian dispersion σ_s of the surface field, a small dispersion σ_b on the interior field due to residual effects of particle size (which in this model is a second order effect), and the fraction p of surface cations relative to the total number of cations.

To compare both models we analysed a series of α -FeOOH spectra with mean particle diameter 350 Å, as determined from X-ray diffraction, for temperature between 77 K and 270 K.

When using our model, satisfactory fittings could be obtained throughout the temperature range a constant value σ_b a value σ_s and p increasing with increasing temperature which is a logical evolution, and with fitted maximum field values H_i^0 which correspond very well with the values obtained for macrocrystalline goethite /4/.

When using the Mørup model, with a logarithmic normal distribution, characterized by a size parameter v_0 and a dispersion parameter σ , a good fit was also obtained and realistic values were also found for the extrapolated maximum internal fields H_i . However it was necessary to change the dispersion parameter significantly through the temperature range, which is fundamentally inconsistent with the supposition that σ is a fixed geometrical distribution parameter.

As a conclusion we can say that both Mørups model and ours seem to work equally well at any chosen temperature, but when temperature dependence is considered it seems that the size distribution effect is of second order compared to the surface effects.

References

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