Research Article

Comment on Phys. Rev. Lett. "All-Electron Self-Consistent GW Approximation: Application to Si, MnO, and NiO". Magnetic moment of NiO

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By this Comment we would like to express our deep scepticism about "Excellent agreement with experiment for many properties" of NiO claimed in the abstract of a paper in Phys. Rev. Lett. **93**, 126406 (2004) by Faleev et al. [1] which has been obtained with "a new kind of self-consistent GW (SCGW) approximation based on the all-electron, full potential linear muffin-tin orbital method."

This excellent agreement in the SCGW approach is based, among others but this can be verified, on the obtained value of 1.72 μ_B for the Ni magnetic moment. This value is indeed improved, in comparison to 1.28 μ_B obtained within the LDA, becoming closer to an experimental value of 1.9 μ_B. However, we claim that this experimental value is presently documented to be wrong, is too small, though this value has often been quoted in literature in last 30 years. The literature for the 1.9 μ_B value has not been given in the commented paper [1] but likely it is quoted after Ref. 2 from 1983. Earlier experimental papers have provided 1.64 μ_B in 1962 and 1.77 μ_B in 1968. Here we would like to put

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attention that the recent very detailed experiment of the Grenoble group from 1998 has provided 2.2±0.3 μ_B at 300 K [3]. This experimental finding has been recalled already in Refs 4–8. We made use of this value in discussion of our calculations for the spin and orbital moment in NiO in a Letter to Phys. Rev. Lett. (LF7313, submitted 8.06.1999) [6, 7]. Our point of the Comment is that even accepting the agreement between the calculated value of 1.72 μ_B and experiment of 1.9 μ_B, but never excellent, the calculated value 1.72 μ_B is substantially too small with respect to the real value in NiO. The disagreement becomes larger if one realizes that the experimental value of 2.2 μ_B was derived at 300 K. Extrapolation of this value to the zero temperature by means of a well-known equation m(T)/m(0)= $[(1-(T/T_N)^2]^{1/2}$ leads to 2.6 μ_B at T = 0 K. In such circumstances the claimed "excellent agreement" is not at all justified. The acceptance by Editors and referees of Phys. Rev. Lett. of this excellent agreement (10%, what should we write in case of 1-2% agreement?) would indicate their conviction that the understanding of the magnetism and electronic structure of NiO has been finally solved or at least largely improved. By expressing our scepticism we would like to say that it is not at all the case. It is a bad overlook of the Editor and referees allowing for the neglect of all works of the Grenoble group on NiO.

We would like to put attention to our atomistic approach to NiO based on an assumption that the paramagnetic atom/ion preserves largely its integrity also in a solid [9–11]. We start analysis of NiO from the detailed analysis of the single-ion effects like the low-energy electronic structure of the Ni^{2+} ion [6, 8]. Within the Quantum Atomistic Solid State theory (QUASST), taking into account strong electron correlations[12], basically of the on-site origin, the intraatomic spin-orbit coupling, crystal-field interactions, yielding discrete energy states, completed with inter-site spin-dependent interactions we have calculated the Ni magnetic moment at T = 0 K as 2.54 μ_B, i.e. really very close to the experimental value. Moreover, we have calculated the orbital and spin contributions to the magnetic moment as well as physically adequate thermodynamics [6, 10, 12]. We have managed to describe in the consistent way both the insulating state, the paramagnetic and magnetic state with the description of, for instance, the λ-peak at T_N in the temperature dependence of the heat capacity [10, 13].

In conclusion, we claim that any approach neglecting the spin-orbit coupling and the orbital magnetism is not physically adequate to 3*d* oxides and that in reaching "excellent agreement" too small experimental value of 1.9 μ_B instead of 2.2 μ_B, or even 2.6 μ_B, has been taken for

the Ni magnetic moment in NiO.

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Added 20.02.2007. This Comment has been written in an abnormal situation in the Phys. Rev. Lett. Office unabling publication of results on the magnetism, electronic structure and the orbital magnetism within the classical many-electron crystal-field approach on NiO and other 3*d* oxides whereas papers within other theories, even without any relevance to experimental observations are often published. We challenge referees of Phys. Rev. Lett. being in full respect to them as the best referees - but we also know the scientific importance of our results. The publication of Comment should be the scientific obligation of each journal claiming to be a scientific journal.