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Hartree-Fock calculations for $FeCl_2$ and $FeBr_2$: The question of the ^{57m}Fe quadrupole moment

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Two previous theoretical evaluations of the ^{57m}Fe quadrupole moment (Q), based on different formalisms—namely, the Hartree-Fock method and the linearized augmented-plane-wave procedure using the local density approximation—have yielded values of Q of 0.082 and 0.16 b, respectively, differing by almost a factor of 2. In both cases, $Q(^{57m}\mathrm{Fe})$ was obtained from experimental quadrupole coupling constants through investigation of electric field gradients (EFG's) at the ^{57m}Fe site. The present work reexamines the earlier Hartree-Fock approach through a careful analysis of the influence of the sizes of the basis sets used, inclusion of the rare-gas environment of the ^{57m}Fe probe in the Mössbauer experiments, and incorporation of electron-electron correlation effects on the EFG's, favoring a value for $Q(^{57m}\mathrm{Fe})$ of 0.110 (0.005) b.

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I. INTRODUCTION

In Mössbauer experiments, no nuclear probe has been used more frequently than the isotope ^{57m}Fe. Extraction of information about the electronic environment of the ^{57m}Fe probe from quadrupole interaction data requires knowledge of the ^{57m}Fe quadrupole moment $Q(^{57m}\mathrm{Fe})$. This quantity, however, has been the subject of a long-standing controversy. While it had been previously estimated to be in the range of 0.15–0.28 b,[1,2] a Hartree-Fock calculation by Duff, Mishra, and Das[3] yielded a substantially smaller value $Q(^{57m}\mathrm{Fe})=0.082$ b. The authors analyzed the results of Mössbauer experiments[4,5] in which $\mathrm{Fe}X_2$ ($X=\mathrm{Cl},\mathrm{Br}$) molecules embedded in a solid argon matrix had been investigated. The value of $Q(^{57m}\mathrm{Fe})$ was deduced from measured quadrupole interaction frequencies in conjunction with the calculated electric field gradients (EFG's) as found at the ^{57m}Fe site. The small value of $Q(^{57m}\mathrm{Fe})$ resulting from this treatment was subsequently confirmed[6] by the experimentally determined ratio of the quadrupole coupling constants (e^2qQ) for ^{54m}Fe and ^{57m}Fe in the same iron compounds in conjunction with nuclear theory applied to the high-spin (10^+) isomer ^{54}Fe.[7]

The most recent theoretical effort[8] directed at the evaluation of the ^{57m}Fe quadrupole moment contradicts the earlier work,[3] yielding a value of $Q(^{57m}\mathrm{Fe})=0.16$ b. This reassessment is based on the interpretation of experimental quadrupole splitting data in a number of compounds, using the linearized augmented-plane-wave (LAPW) band structure[8,9] method based on the local density approximation (LDA). Thus results obtained from two different theoretical procedures are seen to be in obvious conflict, which requires resolution.

The work presented here is an extensive improvement over the former Hartree-Fock approach toward solution of the $Q(^{57m}\mathrm{Fe})$ problem and, particularly, at a careful reexamination of the hypotheses underlying the earlier work.[3] In this way, it is hoped that a reconciliation between the two differing views[3,4] may be achieved eventually.

The special emphasis on the present calculation can be summarized as follows.

(1) The choice of suitable basis sets: namely, basis sets which can be expected to describe local electronic properties reliably, of particular relevance being the basis set used for Fe in the solid-state calculations.

(2) Incorporation of possible effects induced by the solid Ar matrix surrounding the probe molecules.

(3) Inclusion of electron-electron correlation effects.

Neither (2) nor (3) were taken into account in the earlier Hartree-Fock calculation,[3] where the units $\mathrm{Fe}X_2$ ($X=\mathrm{Cl},\mathrm{Br}$) were treated as free molecules based on the assumption of insignificant interactions with the rare-gas environment, and the influence of electronic correlation on the EFG's was not considered.

II. METHODOLOGY

As demonstrated in several previous theoretical investigations on $FeCl_2$ and $FeBr_2$, and most explicitly in Refs. 10 and 11, the appropriate treatment of these system with the aim to evaluate the electric field gradient V_{zz} at the nuclear site of Fe is an intricate problem in view of two major complications: (1) V_{zz} depends very sensitively on the distance between Fe and Cl(Br), and (2) the individual contributions to the net value of V_{zz} cancel to a large extent. Both situations necessitate a very careful description of the electronic system of the selected molecules, most importantly the one of the Fe

TABLE I. Comparison of $\langle 1/r^3 \rangle$ at the nuclear site of Fe as obtained from both Hartree-Fock and LDA computations using the (7/6/5/1) Fe basis set chosen in this work and from numerical calculations.

Quantity	Atomic HF calculation $[e/a_0^3]$	Atomic LDA calculation $[e/a_0^3]$	Standard value[a] $[e/a_0^3]$
$\langle 1/r^3 \rangle$ $(2p)$	468.26	470.00	468.58
$\langle 1/r^3 \rangle$ $(3p)$	52.04	53.50	55.71
$\langle 1/r^3 \rangle$ $(3d)$	4.93	4.55	4.86

[a] Reference 13.

atom, which underscores the relevance of an adequate choice of basis sets.

For the Fe basis set selection, the following criterion was adopted. This basis set is required to give an adequate representation of wave-function-related properties of the Fe atom. The most important one of these is, in the context of the problem under study, the expectation value $\langle 1/r^3 \rangle$ at the nuclear site of the atom, which, in the framework of the Hartree-Fock approach, is for each atomic orbital directly proportional to the V_{zz} component of the EFG tensor associated with that orbital. We chose a (7/6/5/1) basis set[12] (consisting of 7 *s*, 6 *p*, 5 *d*, and 1 *f* functions), comparing the $\langle 1/r^3 \rangle$ expectation values derived for the Fe $2p$, $3p$, and $3d$ shells to standard numerical values for these quantities in the neutral atom.[13] Particular emphasis was placed on a satisfactory reproduction of the $3d$ contribution. From Table I, it is obvious that the basis set chosen reproduces both $2p$ and $3d$ contributions to V_{zz}. The somewhat larger deviation found for the $3p$ shell can be tolerated since the effect of the Fe *p* shells on the EFG at the nuclear site of Fe is expected to be small in comparison to the Fe $3d$ shell.[3]

For comparison with the Hartree-Fock approach, we also carried out a computation of the atomic $\langle 1/r^3 \rangle$ expectation values for the Fe atom using an LDA procedure based on the Becke functional.[14] As can be seen from Table I, by comparison with the numerical value for the Hartree-Fock approach,[13] the $3d$ contribution is significantly underestimated. Apparently, the LDA procedure can therefore decrease the EFG at the nuclear site of the Fe atom and consequently could lead to an overestimation of the ^{57m}Fe quadrupole moment. Similar considerations as for Fe were followed in selecting basis sets for Cl (Ref. 15) and Br (Ref. 16) in the Hartree-Fock investigation of $FeCl_2$ and $FeBr_2$. All computations were carried out using the program GAUSSIAN 98.[17]

III. RESULTS AND DISCUSSION

In an initial series of calculations, aimed at a test for the adequacy of our approach, we computed the ratio ρ of the experimental quadrupole interaction frequencies observed for $FeBr_2$ and $FeCl_2$ which equals the ratio of the actual V_{zz} components at the nuclear site of the Fe atom for $FeBr_2$ and $FeCl_2$, respectively. The measured value is $R(\text{expt}) = 1.36$ (0.04).[4,5] A sequence of three calculations was carried out, using basis sets of increasing complexity, as indicated in Table II, which contains the theoretical and measured values of ρ for both the free molecules and molecules embedded in an Ar environment. It should be noted that the deviation between theory and experiment decreases with increasing basis set complexity. We also observe that inclusion of an *f* orbital into the basis set used for the Fe atom yields a significant improvement of the calculated value.

In all calculations, the measured Fe-Cl(Br) bond distances were used which were determined as 2.17 Å for $FeCl_2$ and 2.307 Å for $FeBr_2$.[18,19] Although for our evaluation of $Q(^{57m}\text{Fe})$ the bond distances of the embedded molecules are of relevance, as opposed to those for the free species, it should be noted that a geometry optimization for $FeCl_2$ ($FeBr_2$) at the unrestricted Hartree-Fock (UHF) level using the most complex of the basis sets indicated in Table II yields an Fe-Cl (Fe-Br) bond distance of 2.173 Å (2.303 Å), in agreement with experiment. We emphasize that both $FeCl_2$ and $FeBr_2$ result as linear species from these optimizations.

As suggested by Hund's rule, a quintuplet spin state ($S = 2$) was assumed for the two molecules in all of these calculations. However, in view of the strong dependence of the EFG on the spin state of the system, it is essential to test this assumption. Thus a comparative study of total energies of the $FeCl_2$ molecule for spin-singlet, -triplet, and -quintuplet states was carried out. These results show (Table III) a strong preference for the quintuplet spin state, which clearly exhibits the lowest energy in this series. The spin contamination, not exceeding 0.3%, was found to be consistently small in this series of computations.

For both $FeBr_2$ and $FeCl_2$ we investigated the contribu-

TABLE II. Values of the V_{zz} component in units of e/a_0^3 for free and Ar embedded $FeCl_2$ and $FeBr_2$ molecules as obtained for a sequence of basis sets.

Basis set for Fe	(7/6/4)	(7/6/5)	(7/6/5/1)	(7/6/5/1)	Experiment[a]
$V_{zz}(FeCl_2)$	0.598	0.596	0.578		
$V_{zz}(FeBr_2)$	0.902	0.796	0.791		
$V_{zz}(FeCl_2Ar_{12})$				0.572	
$V_{zz}(FeBr_2Ar_{12})$				0.778	
ρ[b]	1.508	1.334	1.368	1.360	1.36(0.04)

[a] Reference 5.

[b] ρ represents the ratio of the EFG's for the $FeCl_2$ and $FeBr_2$ systems.

TABLE III. Comparison of total energies of different spin states for $FeCl_2$.

Spin state	Total Energy of $FeCl_2$ [keV]
Singlet	−59.319
Triplet	−59.317
Quintuplet	−59.326

tions of the individual molecular orbitals to the total EFG's. Table IV lists the most important V_{zz} contributions, arising from the $2p$-like, $3p$-like, and $3d$-like molecular orbitals. As expected, the main differences between the V_{zz} values for $FeBr_2$ and $FeCl_2$ can be attributed to the halogen orbitals and the halogen nuclear charges. Only the Fe $(3p)$ shells appear to be percentagewise significantly influenced by covalency effects, their absolute sizes being quite small.

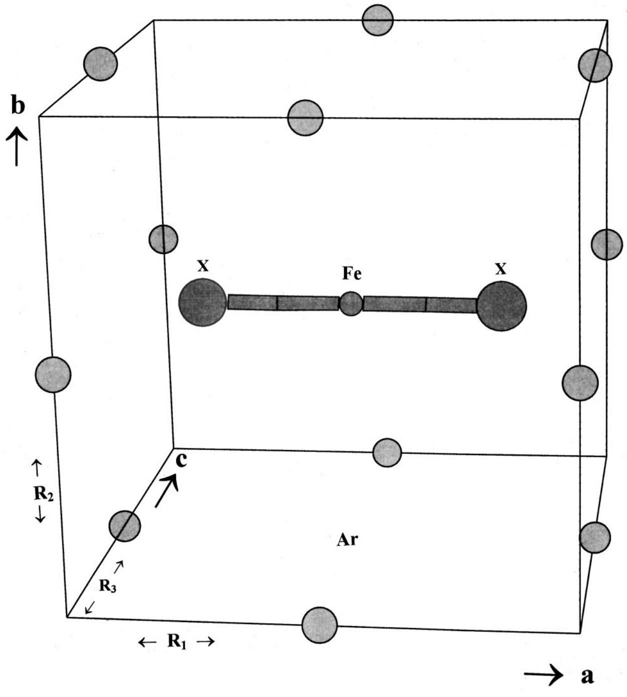

FIG. 1. The FeX_2Ar_{12} model adopted in the calculations described in the text. The three orthogonal axes of the fcc Ar_{12} crystal are denoted by a, b, and c; R_1, R_2, and R_3 refer to the three distortion parameters introduced in the text.

We accounted for the influence of the Ar matrix on the EFG's of $FeBr_2$ and $FeCl_2$ by adopting a cluster model (Table II) involving the molecules surrounded by the 12 nearest Ar atoms. The effect of possible Ar lattice distortion due to the embedded molecule was examined by performing a geometry optimization where the fcc geometry of the undistorted Ar lattice was used for the initial geometry of the FeX_2Ar_{12} ($X = \mathrm{Cl, Br}$) cluster (see Fig. 1), the orientation of the FeX_2 molecule being kept fixed along the a axis, while the Fe-X distance was allowed to vary. Figure 1 indicates the three variable lengths R_1, R_2, and R_3 which describe the Ar_{12} lattice distortion and are related to the lattice parameters a, b, and c. For both $FeCl_2Ar_{12}$ and $FeBr_2Ar_{12}$, the distortion variables R_1, R_2, and R_3 were optimized. As expected, we found $R_2 = R_3$ throughout, so that the FeX_2Ar_{12} structure adopts tetragonal symmetry about the axis of orientation of the FeX_2 molecule. In the case of $FeCl_2Ar_{12}$, optimization resulted in two near-degenerate minima, one with $R_1 > R_2$ ($R_1 = 3.54$ Å, $R_2 = 3.49$ Å) and another one with $R_2 > R_1$ ($R_1 = 3.47$ Å, $R_2 = 3.51$ Å), both differing substantially from the undistorted fcc geometry of the Ar lattice,[20] for which $R_1 = R_2 = 2.66$ Å. The fractional variation in the EFG resulting from the small difference in the geometry for the two minima is found to be in the 10^{-3} range and therefore has little impact on the present discussion. The influence on the EFG due to the Ar environment thus comes out almost the same for the two minima and is included in the results in the fourth column of Table II. The corresponding calculation for $FeBr_2Ar_{12}$, however, yielded only one minimum, which exhibits a considerable degree of distortion with $R_1 = 4.31$ Å and $R_2 = 3.28$ Å. This noticeable contrast between the two systems is reflected by the somewhat larger difference between the EFG's for the free molecule and the solid in case of $FeBr_2$. To test the sensitiveness of the geometry optimization with respect to the orientation of the $FeCl_2$ molecule, the latter was varied systematically in the equatorial plane of the Ar cage. The orientation along the a axis, as indicated in Fig. 1, emerged from this study with highest stability, as documented by the data contained in Table V. We also want to point out that, from our computation, the Ar cage exerts very little influence on the Fe-Cl(Br) bond length, which changes

TABLE IV. The dominant electronic shell contributions to the V_{zz} component in units of (e/a_0^3) for $FeCl_2$ and $FeBr_2$.

	Fe$(2p)$	Fe$(3p)$	Fe$(3d)$	Halogen orbitals	Halogen nuclear charges
$FeCl_2$	0.275	0.066	2.805	−3.493	0.980
$FeBr_2$	0.295	0.104	2.894	−4.105	1.690

TABLE V. Dependence of the total energy of $FeCl_2Ar_{12}$ on the orientation of the $FeCl_2$ molecule in equatorial plane of the Ar_{12} cage (see Fig. 1). The angle Φ denotes the azimuthal angle included by the molecule with the c axis of the cage. Thus the situation shown in Fig. 1 corresponds to $\Phi = 90°$.

Φ [deg]	Energy [eV][a]
50	0.81
60	0.46
70	0.20
80	0.05
90	0.00
100	0.05
110	0.20
120	0.46
130	0.81

[a] The energy minimum of −230.555 [keV] is used as the zero of the energy scale.

TABLE VI. EFG at the Fe site of $FeCl_2$ and $FeCl_2Ar_{12}$ by the Hartree-Fock procedure and various post-Hartree-Fock methods to include many-body effects.

Procedure	System	V_{xx}	V_{yy}	V_{zz}
HF[a]	$FeCl_2$	−0.289	−0.289	0.578
MP2[b]	$FeCl_2$	−0.283	−0.283	0.566
CID[c]	$FeCl_2$	−0.282	−0.282	0.565
CCD[d]	$FeCl_2$	−0.278	−0.278	0.557
MP2[b]	$FeCl_2Ar_{12}$	−0.279	−0.279	0.559

[a] Hartree-Fock.

[b] See Ref. 17.

[c] Configuration interaction with double substitutions, Ref. 17.

[d] Coupled cluster theory with double substitutions, Ref. 17.

by less than 0.4% as compared to the respective bond length of the free molecular species. Thus the Fe-Cl bond length of 2.173 Å found for the free $FeCl_2$ molecule is to be compared with the very slightly reduced Fe-Cl bond length of 2.167 Å in $FeCl_2Ar_{12}$.

From column 4 of Table II, the overall impact of the Ar matrix on the EFG appears to be quite weak for both $FeCl_2$ and $FeBr_2$, amounting to 1% and 2%, respectively. However, the environmental effect is significant in improving an already good agreement with the experimental ratio ρ,[5] as achieved at the level of the purely molecular investigation, to apparently perfect when the Ar cage is included in the physical model.

We want to point out that the impact of a noble-gas environment on the electric field gradient at the nuclear site of Fe in $FeCl_2$ has been previously studied using the Hartree-Fock method by Bominaar *et al.*,[11] who investigated the system $FeCl_2Ne_n$ with $n=2,...,6$ for various geometric configurations of the surrounding Ne atoms. Some fluctuations of the quadrupole interaction with the arrangement of Ne_n were recorded: however, the incomplete representation of the noble-gas atom cage employed in conjunction with the lack of a geometry optimization for $FeCl_2Ne_n$ in Ref. 11 as well as the differences in the noble-gas matrix, basis sets, and Fe-Cl distances used make a direct quantitative comparison of this earlier study with the present work difficult.

The effect of electron-electron correlation on the EFG was taken into account through applications of various post-Hartree-Fock procedures. A many-body perturbation theory (MBPT) treatment[2] at second order (MP2) (Ref. 17) was carried out and compared with results we have obtained by a configuration interaction (CI) (Ref. 17) and a coupled cluster (CC) (Ref. 17) calculation, where double substitutions from the Hartree-Fock determinant were used in both cases. These methods were applied first to the $FeCl_2$ molecule, the EFG results obtained being displayed in Table VI. From these values, the incorporation of many-body effects is seen to lower the EFG as compared to the results from the Hartree-Fock procedure, the reductions in the magnitudes of the EFG slightly increasing with the accuracy of the correlation procedure applied. However, the maximum deviation, as seen from Table VI, from the Hartree-Fock result amounts to less than 4%, so that the impact of electron-electron correlation can be considered to be relatively small for the system $FeCl_2$. We have also included the Ar environment in evaluating many-body effects on the EFG, using the $FeCl_2Ar_{12}$ cluster. We have employed for this purpose the MP2 procedure and found the environmental effect on the many-body contribution only about 1%. Using the EFG result for $FeCl_2Ar_{12}$ from Table VI which includes both many-body and environmental effects and the experimental value of the nuclear quadrupole coupling constant,[4,5] we arrive at a result of 0.110 b for $Q(^{57m}\mathrm{Fe})$. For $FeBr_2$, using the result for the EFG in Table II which includes the environmental effect, but no many-body correction, and the experimental value of e^2qQ for $FeBr_2$ in the Ar lattice, we find $Q(^{57m}\mathrm{Fe}) = 0.107$ b.

To arrive at a confidence limit for $Q(^{57m}\mathrm{Fe})$, we have used the following considerations. First, as far as the dependence of the EFG on the choice of basis set is concerned, our results in Table II indicate that the limit of convergence with respect to the basis set size appears to have been attained. The influence of the environment on the EFG has been found to be rather small, of the order of 1%, and so one does not expect any significant effect from this factor on the confidence limit. The impact of many-body effects has been tested using a number of different methods based on the state-of-the-art computational techniques available and found to be about 4%. The many-body effects, however, depend upon the completeness of the molecular basis set used in the MBPT, CI, and CC procedures, which is difficult to attain from practicability considerations in cluster computations. We can therefore ascribe quite conservatively a 50% confidence limit to the many-body contribution to the EFG, leading to a corresponding confidence limit of 0.002 b for $Q(^{57m}\mathrm{Fe})$. Considering next the difference between the values of Q from $FeCl_2$ and $FeBr_2$, we can add another 0.002 b and round off the net confidence limit at 0.005 b, leading to a result of $Q(^{57m}\mathrm{Fe}) = 0.110$ (0.005) b from the present investigations.

IV. CONCLUSION

In conclusion, the EFG's at the ^{57m}Fe nuclear sites in $FeCl_2$ and $FeBr_2$ in the Ar lattice have been reevaluated using the Hartree-Fock method along with several post-Hartree-Fock procedures to include the influence of many-body effects. The systems chosen are among the best available systems for accurate Hartree-Fock investigation, because they involve relatively small, almost isolated, molecular units. Among the various factors examined for their influence on the EFG, the choice of the basis set is found to be the principal one. The influence of the Ar lattice environment was included by considering a FeX_2Ar_{12} ($X = \mathrm{Cl, Br}$) cluster and performing full Hartree-Fock optimizations on these units. From this investigation, the influence of the rare-gas environment was found to be only of the order of 1%. The influence of many-body effects was determined to be somewhat more important than environmental effects, amounting to about 4%.

The value of the $Q(^{57m}\mathrm{Fe})$ emerging from our recent investigations of 0.110 (0.005) b is about 20% higher than the earlier Hartree-Fock result for the same molecular systems and is in good agreement with the results of perturbed angular correlation (PAC) and Mössbauer measurements[6] on ^{54m}Fe and ^{57m}Fe, respectively, in the same iron compounds combined with nuclear theory[7] for the high-spin ^{54m}Fe isomer. The LDA-based value of the $Q(^{57m}\mathrm{Fe})$ of 0.16 b from the LAPW band structure method,[8] while in better agreement with the present finding than the earlier Hartree-Fock value, is still about 45% higher. The comparison between Hartree-Fock and LDA expectation values of $\langle 1/r^3 \rangle$ for the Fe atom in Table I suggests a 10% reduction in the LAPW result for ^{57m}Fe, which would reduce it to 0.144 b. It is suggested that a further refinement of the LAPW approach including a careful study of convergence tests on the basis sets used and the influence of many-body effects, as carried out here, could contribute to the understanding of the remaining discrepancy with the present value of $Q(^{57m}\mathrm{Fe})$. Additionally, it would be helpful for comparison with the present work to conduct supercell APW investigations of the EFG in the systems $FeCl_2$ and $FeBr_2$ in the Ar lattice studied here.

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