Reply to "Pressure dependence of the hyperfine field in Eu intermetallics"

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Devine has suggested an alternative solution to explain the pressure dependence of the effective hyperfine field in Eu intermetallics. We show that the analysis of Devine is based on assumptions which are very difficult to justify. Additional comments on the quantitative analysis are presented.

In a recent paper of the authors[1] it was shown that the volume dependence of the effective hyperfine field (B_{eff}) in Eu intermetallics is mainly caused by the volume change of the contribution from the conduction-electron polarization of the neighboring magnetic ions (B_{NN}). In contrast to this analysis, Devine[2] proposed a different interpretation assuming that the contribution of the conduction-electron polarization of the magnetic ion itself (B_{sp}) is the main source for the observed pressure dependence of B_{eff}. Although the underlying assumptions[2] are difficult to verify, the authors want to show that these assumptions even contradict one's expectation.

Devine starts with a magnetization at the origin, which is surrounded by the magnetic atoms with spin $\vec{S}(\vec{R}_\alpha)$, $\vec{R}_\alpha$ being the position of the atoms. Applying linear response theory, the polarization of the atom at the origin is given by

$$\sigma(0)=\sum_{\alpha,n}\frac{J_n}{g^2\mu_B^2}\chi_n(\vec{R}_\alpha)\vec{S}(\vec{R}_\alpha)=\sum_n\sigma_n(0) \quad .$$

Of course, this is the most general form of the linear response to magnetic atoms with spins $\vec{S}(\vec{R}_\alpha)$ if the spatial dependent susceptibility $\chi_n(\vec{R}_\alpha)$ [or the correlation function $J_n\chi_n(\vec{R}_\alpha)$] includes every mixture of s electrons with p, d, f, etc. electrons. Dividing σ_n into an intra-atomic and an interatomic part, we have

$$\sigma^{(1)}(0)=\frac{J_n}{g^2\mu_B^2}\chi_n(\vec{R}_\alpha=0)S(0) \quad ,$$

$$\sigma^{(2)}(0)=\sum_{\substack{\alpha\\ \vec{R}_\alpha\neq 0}}\frac{J_n}{g^2\mu_B^2}\chi_n(\vec{R}_\alpha)S(\vec{R}_\alpha) \quad .$$

Devine now concludes, that the "intra-atomic" susceptibility and the "interatomic" susceptibility are both proportional to the bulk susceptibility $\sum_{\vec{R}_\alpha}\chi_n(\vec{R}_\alpha)$,

$$\chi_n(\vec{R}_\alpha=0)\sim\chi_n(\vec{R}_\alpha)\sim\sum_{\vec{R}_\alpha}\chi_n(R_\alpha) \quad ,$$

i.e., proportional to the partial density of states at the Fermi level $\rho_n(E_F)$, apart from factors independent of the volume. From this conclusion it is quite obvious that the pressure dependence of $\sigma_n^{(1)}(0)$ could be of similar magnitude as the pressure dependence of $\sigma_n^{(2)}(0)$. However, this conclusion is difficult to justify.

In $\chi(\vec{R}_\alpha=0)$ and $\chi_n(\vec{R}_\alpha\neq 0)$ are "intra-atomic" and "interatomic" correlation functions, respectively, which should depend on volume in a different way; for instance, even for large distances between the spins $\vec{S}(\vec{R}_\alpha)$, $J_n\chi_n(\vec{R}_\alpha=0)$ should not vanish. In contrast, $J_n\chi_n(\vec{R}_\alpha)$ must vanish for very large distances, i.e., $J_n\chi_n(\vec{R}_\alpha\neq 0)$ cannot exhibit the same volume dependence as $J_n\chi_n(R_\alpha=0)$. This shows that the analysis of Devine is based on assumptions which are very difficult to justify. Our conclusion is supported by the available experimental results[1,3]: According to Devine's model, the relative contributions of s and non-s conduction electrons should be simply reflected in *both* B_{sp} and B_{NN}, only with a difference in the sign. Experimentally,[3] it has been found that compounds like $EuPt_2$ and $EuZn_2$ which have positive B_{NN} values show a dominant s-conduction electron contribution to B_{NN}, where as in all other compounds with negative B_{NN} values the non-s conduction electron contributions to B_{NN} are

dominant. B_{sp}, on the other hand, is always governed by the s contribution (positive for all Eu compounds). Thus, the s and non-s parts of the conduction electrons do not contribute in the same way in B_{sp} and B_{NN}. Furthermore, we do not see reasons to consider the pressure effect on B_{NN} to be nondominant since we have shown experimentially [see Ref. 1 and Fig. 7(a)] that for $Eu_{0.5}Yb_{0.5}Sn_3$ $\partial B_{eff}/\partial p$ decreases by more than 50% compared to $\partial B_{eff}/\partial p$ in $EuSn_3$. In both systems B_{sp} is about the same, however, B_{NN} is reduced by a factor 2 in $Eu_{0.5}Yb_{0.5}Sn_3$ compared to $EuSn_3$ which shows cleanly that B_{NN} is mainly responsible for the observed pressure dependence of B_{eff} in this system.

Apart from the above-mentioned arguments, a new interesting point of view is brought up by Devine: He estimated the pressure effect on B_{sp} caused by the shift of the Fermi energy E_F due to the volume change. This effect results in the case of $LaSn_3$ in an increase of $\rho_d(E_F)$ and thus in a change of B_{sp} which is of the order of magnitude of the measured values. However, the calculated change in B_{sp} is very sensitive to details in the band structure. For example, band-structure calculations of $EuCu_5$ (Ref. 4) show that the slope of $\rho_d(E_F)$ at E_F is five times smaller than for $LaSn_3$. Consequently the value of $\partial B_{sp}/\partial p$ is factor 5 smaller than for $LaSn_3$, although both systems do not show large differences in $\partial B_{eff}/\partial p$.[1] In addition, the total change in $\rho_d(E_F)$ will be generally reduced due to the broading of the d band under pressure. Furthermore, a negative value for $\partial\rho_d(E_F)/\partial p$ in the case of both $EuPt_2$ and $EuZn_2$ is proposed by Devine. This last proposal is essentially needed for the interpretation of our experimental results and requires experimental or theoretical supports.

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