Spin Scattering Cross Sections for Ag, Au, and Gd in Al

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The spin-orbit-induced spin-flip scattering cross sections for silver, gold, and gadolinium in aluminium are calculated using a simple theory. Reasonable agreement is found between theoretical and experimental results.

Recent paramagnetic-resonance measurements[1] on heavy rare-earth ions dissolved in aluminum revealed "bottleneck" behavior[2] in the case of gadolinium. The bottleneck requires that the relaxation rate for the conduction electrons to the lattice be smaller than the "back" relaxation rate, the so-called Overhauser rate, to the localized spin. Such an imbalance can be varied by increasing the relaxation rate to the lattice by adding, for example, nonmagnetic impurities.[3] In that case the spin-orbit coupling associated with the impurity is the mechanism of the increase. Alternatively, the concentration of the localized impurities may be reduced so that the back relaxation from the conduction electrons to the ions diminishes and enables the conduction-electron lattice relaxation to dominate. The removal of the bottleneck by this method has been clearly demonstrated recently.[1,4]

From the rate of removal of the bottleneck by addition of nonmagnetic impurities or reduction in the number of local-moment spins, one can estimate the spin-flip scattering cross sections for the impurities and the localized ions in the host metal. Both of these aspects have been studied and the cross sections for Ag, Au, and Gd in Al deduced.[1] The purpose of this note is to report calculations of the cross sections using a model for spin scattering in alloys[5] and to compare them with the experimental values taken from Ref. 1. The full expression for the relaxation rate of conduction electrons in a binary alloy has been given in Ref. 5 [Eq. (19)]. Assuming a dilute-alloy limit (which is valid for the case of the concentrations studied experimentally) the rate of change of the relaxation rate with concentration of impurity simplifies to

$$\left.\frac{\partial(1/T_1)}{\partial c}\right|_{c\to 0}=\frac{4(2\pi)^3\rho(E_F)d}{C_{k_F}^2}\,(s_1^I-s_1^{\mathrm{Al}})\times(s_1^I-s_1^{\mathrm{Al}}+2Ss_1^{\mathrm{Al}}),\qquad(1)$$

where the terms have been defined in Ref. 5. Here we have assumed a substitutional model for the impurities and taken the three associated binary-alloy structure factors occurring in the term S to be equivalent and equal to the pure-host-metal structure factor. Furthermore, we have taken only the scattering from p-orbital terms[6] into account, since calculation of the d and f terms for Ag, Au, and Gd revealed these terms to be approximately two orders of magnitude smaller. For very low temperatures (~4 °K) the structure-factor term S becomes extremely small, so that we may finally write the spin-flip cross section as

$$\sigma=\frac{1}{N_0v_F}\,\frac{\partial(1/T_1)}{\partial c}=\frac{4(2\pi)^3\rho(E_F)d}{N_0v_FC_{k_F}^2}\times(s_1^{I2}-2s_1^Is_1^{\mathrm{Al}}+s_1^{\mathrm{Al}2}).\qquad(2)$$

Using Hartree–Fock wave functions and self-consistent potentials[7] we have calculated the relevant scattering matrices (s_1^I for the impurity and s_1^{Al} for aluminium as the host) and evaluated the theoretical spin-flip cross sections using Eq. (2). For Gd the problem was not so clear, because with this orthogonalized-plane-wave (OPW) method the means of taking into account the partly filled nature of an inner core shell is not obvious. However, when calculations were performed either orthogonalizing to the half-filled core shell or leaving it unoccupied, the spin-orbit scattering from the outermost filled $5p$ orbital still dominated. The results of the spin-flip scattering calculations are given in Table I together with the experimental results from Ref. 1. It can be seen that in all cases the theoretical results are reasonably close

TABLE I. Experimental (Ref. 1) and theoretical spin-flip scattering cross sections for impurities in Al.

Impurity	$10^{18}\sigma_{\text{theoret.}}$ cm^2	$10^{18}\sigma_{\text{expt.}}$ cm^2
Ag	1.1	1.5 ± 0.7
Au	12.6	6 ± 3
Gd	7.8	12 ± 6

(approximately within the limits of experimental error) to the experimental values. For Gd it is possible that the 5*d*-orbital character should be present in the conduction-electron wave function near the rare-earth ion,[8] in which case the spin-orbit scattering of the electrons would be enhanced, consistent with the observed discrepancy between theory and experiment. This effect can be treated by admixing 5*d* character into the OPW, but the magnitude of the admixture parameter would remain essentially an adjustable parameter.

In the calculations presented we have not made allowance for screening effects, even though these must be present in the case of Ag and Au, since this is not easily done in the framework of the OPW method. However, some idea of the effect may be obtained from the results of phase-shift calculations, where the effects of screening can be allowed for or ignored. The spin scattering cross section for cesium in sodium, for example, has been found to be twice as large when screening is ignored as it is when screening is taken into account.[9] Here the effective negative valence difference (– 1.68) arises from atomic volume differences[10] and not from pure valence differences, but it has the same sign and order of magnitude as we would expect for Ag and Au in Al. If the calculated cross sections for Ag and Au should be reduced by a factor of 2 due to the "electron repulsion" in the neighborhood of the ion, the experimental and theoretical results for Au would be better reconciled, but those for Ag would not.

In Ref. 1 comparison was made between the scattering cross sections for Au and Ag in Al and those in Li and Na,[6] but it was noted that in the latter two cases the cross sections were five to ten times larger than in Al. Such a large difference cannot be explained on the basis of a simple model, as is usually assumed, where scattering produced by an impurity in different hosts is proportional to the difference in spin-orbit scattering between the host and the impurity. Furthermore, on the basis of the previous discussion, it seems unreasonable to try to reconcile factors of between 5 and 10 difference between Na and Li as hosts and Al as a host on the basis of screening. What is more likely is that the scattering produced by Ag and Au in Na and Li is anomalously large for other reasons.[11,12]

In conclusion, we believe that the spin-flip scattering results for Ag, Au, and Gd in Al given in Ref. 1 can be reasonably accounted for using a simple model in which spin-orbit coupling associated with the impurity increases the scattering of the conduction electrons.

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[1]C. Rettori, D. Davidov, R. Orbach, E. P. Chock, and B. Ricks, Phys. Rev. B **7**, 1 (1973).

[2]H. Hasegawa, Prog. Theor. Phys. **21**, 483 (1959).

[3]Magnetic impurities clearly also couple the electrons more strongly to the lattice (also via spin-orbit scattering) but in using them to relieve the bottleneck one must admit the possibility of adding other relaxation mechanisms for the localized ions, such as direct exchange between the two magnetic species.

[4]G. Weimann, B. Elschner, K. H. J. Bushcow, and R. P. van Stapele, Z. Physik (to be published).

[5]J. S. Helman and R. A. B. Devine, Phys. Rev. B **4**, 1153 (1971); *ibid.* **4**. 1156 (1971).

[6]J. R. Asik, M. A. Ball, and C. P. Slichter, Phys. Rev. **181**, 645 (1969).

[7]F. Herman and S. Skillman, *Atomic Structure Calculations* (Prentice-Hall, Englewood Cliffs, N. J., 1963).

[8]J. M. Dixon (private communication).

[9]R. A. B. Devine, Ph. D. thesis (University of Warwick, Warwick, England, 1970) (unpublished).

[10]F. J. Blatt, Phys. Rev. **108**, 285 (1957).

[11]J. A. Rigert and C. P. Flynn, Phys. Rev. Lett. **26**, 1177 (1971).

[12]The conduction electrons in alloys with Al as host have a large Fermi energy which is far above the energy of the relevant *p* or *d* "resonance" levels of the impurities. This could account for the reasonable agreement obtained with the simple theory.