Electron energy relaxation by phonons in the Kondo condensate

L. J. Taskinen and I. J. Maasilta
Nanoscience Center, Department of Physics, P.O. Box 35, FIN-40014 University of Jyväskylä, Finland
(Dated: March 23, 2022)

We have used normal metal-insulator-superconductor tunnel junctions as thermometers at sub-Kelvin temperatures to study the electron-phonon (e-p) interaction in thin Aluminum films doped with Manganese, as a function of Manganese concentration. Mn in Al is known to be a Kondo impurity with extremely high Kondo temperature \( T_K \approx 500 \) K, thus our results probe the e-p coupling in the fully spin compensated, unitary limit. The temperature dependence of the e-p interaction is consistent with the existing theory for disordered metals, however full theory including the Kondo effect has not been worked out yet. The strength of the interaction decreases with increasing Manganese concentration, providing a means to improve sensitivity of detectors and efficiency of solid state coolers.

PACS numbers: 73.23.-b, 72.10.Di, 72.15.Qm, 74.50.+r

Recently, there has been a renaissance of the many body physics of magnetic impurities interacting with conduction electrons in metals, known as the Kondo effect [1]. In particular, a lot of understanding has emerged on how the magnetic impurities affect the dephasing [2] and energy relaxation [3, 4] of electrons at low temperatures, by a mechanism where the electron-electron (e-e) interaction is mediated by the exchange interaction with the magnetic impurities [5, 6, 7]. Although this new channel for the e-e interaction is clearly dominant for dephasing [2], it will disappear in the limit where \( T << T_K \). In this extreme unitary limit, the impurity spin will be completely screened by the electrons, with only scatter electrons elastically, and Fermi-liquid theory is know to be valid, if the number of orbital channels \( k \) of the impurity equals twice the spin, \( k = 2S \) [1, 6, 8].

In light of the above, we have decided to study an impurity-host system with an extremely large Kondo temperature \( T_K \approx 500 \) K, Aluminum doped with Manganese [1]. The expected magnetic ion in this system is Mn\(^{2+}\), and if Hund’s rule applies, it has five spin aligned d-electrons, thus \( k = 2S = 5 \). As the Kondo impurity mediated mechanism is then suppressed at our experimental sub-Kelvin temperature range, only direct e-e interaction and electron-phonon (e-p) interaction remain as the main mechanisms for electron energy loss. Here, we concentrate on studying the e-p interaction, which is typically the dominant energy loss channel for the electron system as a whole. This means it is very critical for the performance of bolometric detectors and solid state coolers [10]. In fact, AlMn has already been used in novel NIS bolometers [11], TES microcalorimeters [12] and thin-film solid-state refrigerators [13].

In this work, we present the first experimental results on the e-p interaction in the AlMn Kondo-system. No previous data exist in the literature, to our knowl-

edge. We have measured the temperature dependence and strength of the interaction for samples with varying Mn concentration below 1K, using NIS tunnel junction thermometry [14]. Results show that e-p interaction weakens significantly below the value for clean Al thin films [13]. The strength of the effect depends strongly on the concentration of Mn impurities present.

The scattering rate between electrons and acoustic phonons via deformation potential depends strongly on temperature, \( 1/T_{e-p} \sim T^m \), where \( m \) can have values ranging between 2-7 depending on whether the sample is a metal or a semiconductor, the type and level of disorder and dimensionalities of the electron and phonon systems [10, 17, 18]. For disordered 3D metal films in the limit \( ql < 1 \), where \( q \) is the wavevector of the dominant thermal phonons and \( l \) the electron mean free path, theory predicts [14, 20, 21, 22] that if the scatterers are vibrating with the phonons, interference terms lead to suppression of e-p relaxation. Then \( m = 4 \) (in contrast to \( m = 3 \) for pure samples), and the net power flowing from hot electrons of volume \( V \) at a temperature \( T_e \) to phonons at \( T_p \) is

\[
P_{e-p} = \Sigma V(T^n_m - T^n_p),
\]

where \( n = m + 2 = 6 \) and \( \Sigma \) is a sample parameter. This suppression has recently been observed in Cu and Au noble metal films [23], where the simple theory is expected to be correct, in addition to earlier results on Ti and Hf [24]. The effects of the more complex Fermi surface of Al and the inclusion of the Mn Kondo ions are presently not known theoretically.

A Schematic of the samples used to measure the e-p interaction is shown in Fig. 1. We use the hot-electron technique [25] to measure the e-p interaction by overheating the electrons by Joule heat power \( P \) and measuring the resulting temperature \( T_e \). All the measured samples had two long (\( L \sim 0.5 \) mm), electrically isolated Al-Mn normal metal wires separated by 2 \( \mu m \). Since \( L >> L_{e-e} \), the electron-electron scattering length, \( T_e \) is
well defined without complications from non-equilibrium [4]. The upper wire was heated by applying a voltage across a pair of superconducting Al leads in direct metallic contact to AlMn, forming SN junctions. These junctions provide excellent electrical, but very poor thermal conductance due to Andreev reflection, as the junctions are biased within the superconducting gap $\Delta$ of Al in all the data shown here. Thus, due to the lack of outdiffusion of electrons, the input heat is distributed uniformly in the interior of the wire [26, 27]. We measure the electron temperature in the middle of the wire, where two additional Al leads form a NIS tunnel junction pair (SINIS), as a function of input the Joule power $P = IV$, measured in four probe configuration. The purpose of the lower Al-Mn wire, with an additional SINIS thermometer on it, is to give an upper limit to the local $T_p$, as the e-p power flow (Eq. 1) depends on both $T_e$ and $T_p$. As our sample volumes are large, the contribution of thermal photon emission [28] is estimated to be insignificant compared with phonon emission.

Several e-p samples were fabricated in three different Mn concentrations (S1,S2,S3), on nitridized silicon chips using electron beam lithography and three-angle shadow evaporation of Al and Al-Mn [29]. Four probe samples with similar Al-Mn wire dimensions to the e-p samples were also fabricated for accurate resistivity measurements between room temperature and 4K (samples RS1, RS2 and RS3). Evaporation was done from zero angle using the same evaporation parameters (growth rate, new crucible every time and same amount of source material) as for the e-p samples. The oxide layer forming the tunnel barrier was produced by thermal oxidation of Al, resulting in tunneling resistances $R_T \sim 6-15$ k$\Omega$. Widths of the Al-Mn wires varied from 300 to 500 nm, thicknesses from 50 to 60 nm, and length was always $\sim 487 \mu$m. The small dimensions were measured accurately for each sample with an AFM.

The composition of the evaporated AlMn films were determined using elastic recoil detection analysis [30].

Results for samples RS1, RS2 and RS3 show atomic Mn concentrations $c_i$ of $(13.6 \pm 1.0)$ %, $(1.3 \pm 0.2)$ % and $(0.54 \pm 0.10)$ %, respectively. These can be compared with the nominal source concentrations of 2 %, 0.65 % and 0.3 % for RS1-RS3, respectively. In evaporation, the Mn concentration increases due to the larger vapor pressure of Mn compared to that of Al.

Data from resistivity measurements is shown in Fig. 2. Samples RS2 and RS3 exhibit a typical metallic resistiv-
ity, where the phonon contribution dominates at high temperatures (region where \(dp/dT > 0\)), whereas below 30 K the impurity resistivity dominates. However, if one looks at the low temperature region more carefully [Figs. 2(b) and (c)], clear minima for both samples can be seen. This is a signature of the Kondo effect. In sample RS1 with the highest Mn concentration, the contribution of the Kondo resistivity is so large that it completely overwhelms the phonon contribution in the whole temperature range all the way up to room temperature (\(dp/dT < 0\)). The shape of the \(\rho\) vs. \(T\) curve agrees qualitatively with the Kondo theory in the strong coupling limit \(T < T_K\). Quantitative comparison is difficult, as the temperature range is such that only numerical renormalization group calculations are valid [1]. The residual resistivity at 4.2 K is approximately linear in \(c_t\), as expected for the unitary limit [1].

![Electron temperature vs. power density for AlMn e-p samples](image)

**FIG. 4:** Measured electron temperatures of the AlMn e-p samples vs. applied power per unit volume. Circles, squares and triangles denote data from samples S1, S2 and S3, respectively. Continuous line is a guide for the eye \(T^3/6\). Dashed line corresponds to Al data in the pure limit, \(\rho_\text{Al} = 5\), with \(\Sigma = 6 \times 10^3\) calculated for sample 1 in Ref. [15], using a value for Sommerfeld constant \(\gamma = 135\) J/K² m³.

Having established the relevance of Kondo physics in our samples, we discuss next the e-p measurements, performed in a dilution refrigerator at base temperature \(T = 60\) mK. As AlMn NIS junctions have not been used widely in thermometry before, we first have to ensure that they work as expected. Fig. 2 shows a typical current-voltage characteristics of a AlMn/AlOx/Al SINIS junction, with the refrigerator temperature varying between 60-500 mK. The usual suppression of the current at small bias voltages due to the superconducting gap (\(2\Delta/e \sim 0.4\) mV) is seen. As \(T\) is increased, the gap feature is smeared and more current flows. Thus, by current biasing the the junction and measuring its voltage, very sensitive thermometry is possible. Fig. 3 also shows theoretical modeling based on BCS theory and a thermal model introduced in Ref. [13]. This model takes into account the current and heat flows across the junctions calculated from the exact BCS formulas, in addition to cooling by phonons, back emission of heat from the superconductor, and dissipation due to imperfections of the barrier. We found that only at 60 mK was the full thermal model necessary for a good fit (with one free parameter), at all other temperatures a simple BCS theory with a constant \(T\) was sufficient to model the data. This was checked for all Mn concentrations used. Thus, the SINIS junctions could be calibrated using a calibrated RuO thermometer on the sample stage, taking into account noise overheating at the lowest temperatures, as discussed in Refs. [23, 31].

**TABLE I:** Parameters of the e-p samples. the mean free path \(l\) was calculated from the resistivity at \(T = 60\) mK using the Drude formula, \(\Sigma\) was obtained from the fits to the data in Fig. 3. \(\gamma = (1/\tau_{AlMn})/(1/\tau_{Al}) = 6\Sigma T/(5\Sigma_{Al})\).

| Sample | Source | Mn % | \(l\) (nm) | \(\Sigma\) (10³W/K²m³) | \(r(0.1\) K) |
|--------|--------|------|-------------|-----------------|--------------|
| S1     | 2      | 0.2  | 1.1         | 6.0             |
| S2     | 0.65   | 1.9  | 2.5         | 0.27            |
| S3     | 0.3    | 3.2  | 4.2         | 0.46            |

![Numerical logarithmic derivatives of the measured data](image)

**FIG. 5:** Numerical logarithmic derivatives of the measured data in Fig. 4 for samples S1 (a), S2 (b) and S3 (c).

The data for samples S1, S2 and S3 used to obtain the strength of the e-p interaction is shown in Fig. 4. We have plotted the electron temperatures \(T_e\) (measured with SINIS thermometers) as a function of the input power per sample volume applied to the electron gas, in log-log scale. The temperatures of the unheated wires were found to be low enough, so that the condition.
$T_p^0 > T_p^0$ holds, and the data can be analyzed without the direct knowledge of $T_p$. In this case, the slope of the measured $T_e$ vs. $P$ gives directly the value $1/n$, and the constant multiplier the value $\Sigma$. All samples seem to be consistent with $n = 6$, agreeing with similar Cu and Au samples [23]. The saturation of $T_e$ at low $P$ is due to external noise heating [22]. Values of $\Sigma$ obtained from fits fixing $n = 6$ are shown in Table 4. Clearly, the e-p coupling weakens significantly with increasing Mn concentration. Fig. 4 also shows a comparison with the clean limit for Al films, using $n = 5$ and $\Sigma_{AI} = 1.1 \times 10^3 \text{W/K}^2\text{m}^3$, obtained from Ref. 15. Comparing $\Sigma_{AI}$ with our results, we see that the energy relaxation rate $1/\tau$ is suppressed by an order of magnitude at 100 mK for the highest Mn concentration, see Table 4. Comparison with the data measured for Cu and Au wires of approximately the same film thickness using the same technique [23] gives temperature independent suppression factors ranging from 0.02 (AlMn 2\% vs Au) to 0.3 (AlMn 0.3 \% vs Cu). Finally, to obtain a detailed picture of the temperature dependence, we plot the numerical derivatives $d(\log T_e)/d(\log P)$ of the data in Fig. 5. It is clear that $P \sim T^p$ describes the data well over a large range of $P$ for all samples. The deviations at highest heating powers for samples S1 and S3 are presently not understood.

In conclusion, we have obtained clear evidence for suppression of phonon emission from electrons in the extreme Kondo compensated limit $T << T_K$ in Aluminum doped with Manganese. As Kondo impurity mediated e-e scattering is expected to be suppressed in the unitary limit (other possible Kondo impurities such as Cr and Fe, also have a high $T_K > 300$ K in Al), pure e-e scattering should dominate the dephasing at sub-Kelvin temperatures in this material. The observed suppression of e-p scattering also leads to an extension of the temperature range of e-e dephasing, therefore AlMn is a good candidate material to study the e-e interaction. For hot-electron bolometers, a weak e-p coupling leads to increased sensitivity. The expected improvement of the noise equivalent power (NEP $\sim \sqrt{\Sigma}$) for the high concentration AlMn is a factor $\sim 4$ over Cu ($\sim 7$ over Au). Weak e-p coupling is also beneficial for solid state electron coolers.

We acknowledge Tarmo Suppula for help in sample fabrication and Timo Sajavaara and Kenichiro Mizohata for the Mn concentration analysis. We are also grateful to A. C. Hewson and D. E. Prober for valuable discussions. This work was supported by the Academy of Finland under contracts No. 105258 and 205476.

[1] A. C. Hewson, *The Kondo Problem to Heavy Fermions* (Cambridge University press, 1997).
[2] F. Pierre, A. B. Gougam, A. Anthore, H. Pothier, D. Esteve, and N. O. Birge, Phys. Rev. B 68, 085413 (2003).
[3] B. Huard, A. Anthore, N. O. Birge, H. Pothier, and D. Esteve, Phys. Rev. Lett. 95, 036802 (2005).
[4] A. Anthore, F. Pierre, H. Pothier, and D. Esteve, Phys. Rev. Lett. 90, 076806 (2003).
[5] A. Kaminski and L. I. Glazman, Phys. Rev. Lett. 86, 2400 (2001).
[6] M. Garst, P. Wöflle, L. Borda, J. von Delft, and L. Glazman, Phys. Rev. B 72, 205125 (2005).
[7] G. Zaránd, L. Borda, J. von Delft, and N. Andrei, Phys. Rev. Lett. 93, 107204 (2004).
[8] C. Baeuerle, F. Mallet, F. Schopfer, D. Mailly, G. Eska, and L. Saminadayar, Phys. Rev. Lett. 95, 266805 (2005).
[9] P. Nozières and A. Blandin, J. Physique 41, 193 (1980).
[10] F. Giazotto, T. T. Heikilä, A. Luukanen, A. M. Savin, and J. P. Pekola, Rev. Mod. Phys. 78 (2006).
[11] D. R. Schmidt, K. W. Lehnert, A. M. Clark, W. D. Duncan, K. D. Irwin, N. Miller, and J. N. Ullom, Appl. Phys. Lett. 86, 035505 (2005).
[12] S. W. Deiker, W. Doriese, G. C. Hilton, K. D. Irwin, W. H. Rippard, J. N. Ullom, L. R. Vale, S. T. Ruggiero, A. Williams, and B. A. Young, Appl. Phys. Lett. 85, 2137 (2004).
[13] A. M. Clark, A. Williams, S. T. Ruggiero, M. L. van den Berg, and J. N. Ullom, Appl. Phys. Lett. 84, 625 (2004).
[14] J. M. Rowell and D. C. Tsui, Phys. Rev. B 14, 2456 (1976).
[15] P. Santhanam and D. E. Prober, Phys. Rev. B 29, 3733 (1984)
[16] A. Sergeev and V. Mitin, Phys. Rev. B 61, 6041 (2000); Europhys. Lett. 51, 641 (2000).
[17] A. Sergeev, M. Y. Reizer, and V. Mitin, Phys. Rev. Lett. 94, 136602 (2005).
[18] S.-X. Qu, A. N. Cleland, and M. R. Geller, Phys. Rev. B 72, 224301 (2005).
[19] A. B. Pippard, Philos. Magn. 46, 1104 (1955).
[20] A. Schmid, Z. Phys. 259, 421 (1973).
[21] B. L. Altshuler, Zh. Eksp. Teor. Fiz. 75, 1330 (1978), [Sov. Phys. JETP 48, 670 (1978)].
[22] M. Y. Reizer and A. V. Sergeev, Zh. Eksp. Teor. Fiz. 90, 1056 (1986), [Sov. Phys. JETP 63 616 (1986)].
[23] J. T. Karvonen, L. J. Taskinen, and I. J. Maasilta, Phys. Rev. B 72, 012302 (2005).
[24] M. E. Gershenson, D. Dong, T. Sato, B. S. Karasik, and A. V. Sergeev, Appl. Phys. Lett. 79, 2049 (2001).
[25] M. L. Roukes, M. R. Freeman, R. Germain, R. C. Richardson, and M. B. Ketchen, Phys. Rev. Lett. 55, 422 (1985).
[26] Electron temperature is nonuniform within the distance $L_{e-p} = \sqrt{4\tau_{e-p}}$ away from the NS contacts. As $L >> L_{e-p}$ here, no complications arise due to end effects, unlike in an earlier initial work [22].
[27] L. J. Taskinen, J. T. Karvonen, and I. J. Maasilta, Nucl. Instr. and Meth. A 559, 639 (2006).
[28] D. R. Schmidt, R. J. Schoelkopf, and A. N. Cleland, Phys. Rev. Lett. 93, 045901 (2004).
[29] Source materials used for evaporation of Al-Mn films were Goodfellow Al98/Mn2 (atomic %) (S1 and RS1), Al99.35/Mn0.65 (S2 and RS2) and Al99.7/Mn0.3 (S3 and RS3).
[30] M. Putkonen, T. Sajavaara, L. Niinistö, and J. Keinonen, Anal. Bioanal. Chem. 382, 1791 (2005).
[31] J. T. Karvonen, L. J. Taskinen, and I. J. Maasilta, Phys. Stat. Sol. (c) 1, 2799 (2004).