Thermal-Spike Lifetime from Picosecond-Duration Preequilibrium Effects in Hyperfine Magnetic Fields Following Ion Implantation

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The effective hyperfine magnetic fields acting on short-lived excited nuclear states ($27 < \tau < 127$ ps) have been measured for ~7.5 MeV Ir and Pt ions immediately after implantation into iron hosts at room temperature. The observed field strengths decrease with the lifetime of the probe state and are consistent with the hyperfine field being absent for about 6 ps after implantation. As the hyperfine field is quenched while the local temperature exceeds the Curie temperature, these results give a direct measurement of the thermal-spike lifetime. [S0031-9007(99)09022-5]

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When a heavy ion with an energy of several MeV is implanted into a solid it loses energy initially by causing ionization along its path. Once the ion nears the end of its trajectory, however, the energy loss is predominantly through quasielastic atomic collisions which set in motion a cascade of host-atom collisions. The energy density in this collision cascade is so high that a collective "hot spot" or thermal spike is believed to occur for at least a few picoseconds after the ion has come to rest. The thermal spike plays an important [1], but sometimes controversial [2], role in processes such as sputtering, atomic mixing, the clustering of vacancies, and the production of defects. Controversies arise largely because there has been little direct experimental evidence of the thermal spike itself, and model-dependent inferences must be made from measurements of the properties of the modified solid long after the thermal spike is fully quenched. Recently, there has been progress in the theoretical interpretation through developments in molecular-dynamics computer simulations [3,4] which show, among other things, that, in the region around the implanted ion, local melting often occurs and persists for several picoseconds [5,6].

In this Letter we present the first direct measurement of the thermal-spike lifetime for heavy ions implanted into magnetized iron, obtained by studying preequilibrium effects in the hyperfine magnetic fields experienced by the implanted nuclei. Some preliminary aspects and related work have been published in Refs. [7–9]. We used the implantation perturbed angular correlation (IMPAC) technique [7–10]. As this is a time-integral technique, in which the hyperfine fields are sampled throughout the lifetime of the nuclear state, preequilibrium effects in the hyperfine fields will cause the average measured hyperfine fields to depend on the lifetime of the probe state [8].

Beams of 40 MeV ^{16}O from the ANU 14UD Pelletron accelerator were employed to Coulomb excite the low-excitation states of ^{191}Ir, ^{193}Ir, and ^{198}Pt and simultaneously recoil-implant the excited nuclei into an iron host. The target consisted of sequential layers of $^{\mathrm{nat}}$Ir, ^{198}Pt, and $^{\mathrm{nat}}$Ir (0.4, 0.15, and 0.5 mg cm^{-2}, respectively) sputtered onto an annealed Fe foil 1.63 mg cm^{-2} thick, backed by an evaporated 4.5 mg cm^{-2} layer of Cu. These nuclei were chosen because the hyperfine fields for platinum and iridium in iron are large (~10^2 T) and the lowest $5/2^+$ and $7/2^+$ levels in the iridium isotopes have lifetimes that range between 27 and 127 ps. The ^{198}Pt layer provides a comparison with previous work [7].

Backscattered beam ions were registered in an annular counter around the beam axis. Perturbed particle-γ angular correlations were measured by placing a pair of Ge γ-ray detectors at $\pm 115°$ to the beam direction, to serve as monitors, while another pair of detectors was placed in the forward quadrants at a sequence of angles between 0° and $\pm 65°$. To enhance the sensitivity of the measurements to the precessions of the shorter-lived $7/2^+$ states, longer runs were performed with the forward-placed detectors at $\pm 35°$ and $\pm 65°$ to the beam direction. The iron foil was polarized perpendicular to the detector plane and the direction of the polarizing field was reversed frequently. The target assembly was maintained at room temperature throughout the measurements.

In these measurements, the recoiling iridium and platinum ions enter the iron layer of the target with energies between ~5 and ~10 MeV, on average with 7.5 MeV, after spending 0.08 ps in the layers of Ir and Pt. An average recoiling ion stops after ~0.75 ps with a range in Fe of ~0.8 μm (0.6 mg cm^{-2}). Further aspects of the recoil-implantation process have been discussed in Ref. [8].

The implanted nuclei experience the transient hyperfine field B_{tr} while they are in motion and the static hyperfine field B_{st} after they come to rest. Assuming a purely magnetic interaction, and ignoring any preequilibrium effects, the perturbed angular correlation can be written as [8–10]

$$W(\theta, \pm B_{\mathrm{ext}}) = \sum_{k=0,2,4} \frac{b_k \cos[k(\theta \mp \Delta\theta_k \mp \Delta\theta_{\mathrm{tr}})]}{\sqrt{1 + (k\omega\tau)^2}}, \quad (1)$$

where $\Delta\theta_k$ is related to the static-field precession, $\omega\tau$, by $\tan(k\Delta\theta_k) = k\omega\tau$, $\omega\tau = -g\frac{\mu_N}{\hbar}B_{\rm st}\tau$. $\Delta\theta_{\rm tr}$ is the transient-field precession, b_k are the angular-distribution coefficients, $B_{\rm ext}$ is the external polarizing field, g is the g factor of the state of interest, and τ is its mean life. The $\Delta\theta_{\rm tr}$ values applicable in this paper were evaluated from the data in Refs. [8,9,11], which include measurements on the present target with 100 MeV ^{32}S beams.

Figure 1 shows the perturbed angular correlations for the $5/2_1^+ \rightarrow 3/2_1^+$ transitions of energy 129 and 139 keV in ^{191}Ir and ^{193}Ir, respectively. The static-field precession angles, $\omega\tau$, were extracted using a generalization of Eq. (1), which takes account the (small) effects of feeding together with the transient-field precession [12]. The results are summarized in Table I.

The present results for ^{198}Pt agree with several similar IMPAC measurements, reviewed in Ref. [7]. Our precessions for the $5/2_1^+$ states in the iridium isotopes also agree with the only previous IMPAC measurement [19].

To extract the static-field strengths, we adopt mean lives that are weighted averages of recoil-distance measurements [13,14] and values derived from the Coulomb-excitation studies of McGowan *et al.* [16,17] (with which the lifetime measurements agree). The g factors of the states of interest were measured simultaneously, relative to each other, by the transient-field technique [13,14] and normalized to $g(2_1^+)$ in ^{198}Pt [15] (see [8] for some preliminary results) [20].

The effective static-field strengths derived from our IMPAC measurements on the iridium isotopes (Table I) are clearly smaller for the shorter-lived states. These data, from precise, simultaneous measurements of the effective fields for states of differing lifetimes in the same atomic species, give the first unambiguous evidence for picosecond-duration preequilibrium effects in hyperfine fields following implantation [21].

We interpret the observed lifetime dependence of the effective hyperfine field in terms of a preequilibrium quenching of the local magnetization during the collision-cascade and thermal-spike phases of the implantation process. Initially, we assume that the hyperfine field is absent during the thermal spike, after which it rises rapidly to its equilibrium value. Assigning the implantation and thermal-spike processes a total lifetime t_s, the observed hyperfine field in an integral IMPAC measurement, $B_{\rm IMPAC}$, then depends on the lifetime of the probe state as

$$B_{\rm IMPAC} = B_0 e^{-t_s/\tau}, \tag{2}$$

where B_0 is the effective "equilibrium" value of the hyperfine field. As B_0 may include long-lived radiation damage effects, it need not correspond to the field found by other techniques. It is useful to plot $B_{\rm IMPAC}$ as a function of the inverse lifetime (i.e., the decay rate) of the probe state. Our results for the states in ^{191}Ir and ^{193}Ir are plotted in Fig. 2(a) along with the average field obtained from previous radioactivity measurements on the $5/2_1^+$ states [22,23], reevaluated with the present g factors and lifetimes, and plotted at $1/\tau = 0$. The fit of Eq. (2) to the present IMPAC data alone gives $t_s = 7.3 \pm 0.8$ ps and an effective B_0 value that happens to agree with the field obtained in the radioactivity measurements [22,23].

As, in general, there will be considerable subcascade formation following the implantation of heavy ions with energies of several MeV [1], the thermal-spike process would be expected to cause a similar quenching of the hyperfine field for all heavy atomic species implanted into iron under similar conditions. The behavior shown in Fig. 2(a) would then be a global feature of IMPAC measurements on short-lived states ($\tau \lesssim 100$ ps). We have surveyed the literature and, in Figs. 2(b)–2(d), we show those cases of sufficient precision to suggest lifetime-dependent hyperfine fields. Along with the present and previous [7,24,25] results for the Pt isotopes, the data for the rare-earth ions Dy [26], Nd, and Sm [10] are suggestive. While these cases are not compelling in isolation from the present results for iridium, they are all consistent with a

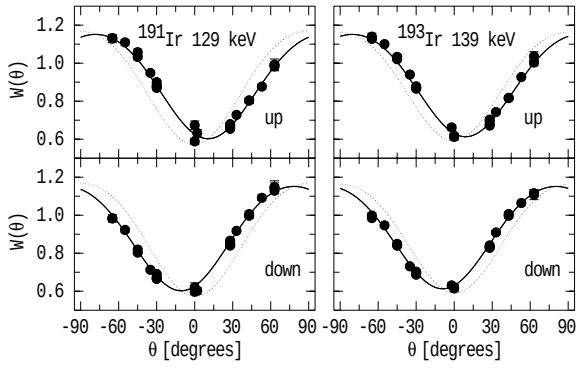

FIG. 1. Perturbed angular correlations for the 129 and 139 keV $5/2_1^+$ states in ^{191}Ir and ^{193}Ir.

TABLE I. Measured precessions and hyperfine fields.

Level	g [a]	τ [b] (ps)	$\Delta\theta_{\rm tr}$ [c] (mrad)	$\omega\tau$ (mrad)	$\lvert B_{\rm IMPAC}\rvert$ (T)
^{191}Ir					
$5/2_1^+$	0.322(20)	128(2)	−8(1)	207(3)	106(7)
$7/2_1^+$	0.401(18)	30(1)	−10(1)	51(2)	89(6)
^{193}Ir					
$5/2_1^+$	0.356(16)	101(2)	−9(1)	177(3)	103(5)
$7/2_1^+$	0.441(16)	27(1)	−11(1)	48(2)	84(5)
^{198}Pt					
2_1^+	0.314(11)	33(2)	−7(1)	45(2)	90(6)

[a] g factors from transient-field measurements [13–15].
[b] Lifetimes from Refs. [13,14,16–18].
[c] Calculated transient-field precessions. See Ref. [9] and text.

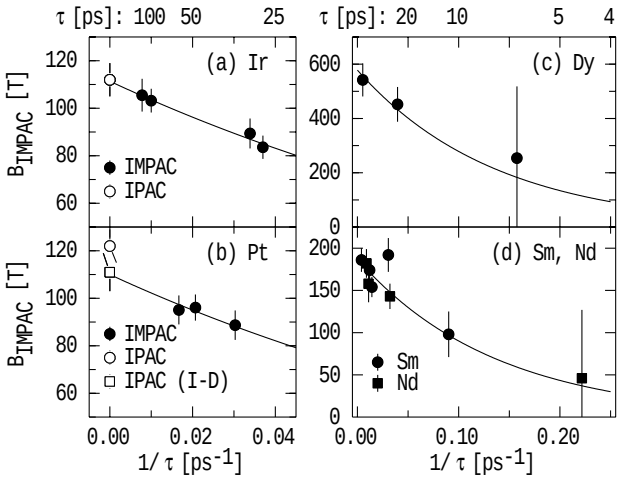

FIG. 2. Static hyperfine magnetic fields from IMPAC measurements plotted as a function of the inverse lifetime (or decay rate) of the probe state. Off-line data from other techniques are plotted at $1/\tau = 0$. The curves correspond to Eq. (2) with $t_s = 7.3$ ps, obtained from fitting the present iridium IMPAC data alone. (a) Present IMPAC and previous radioactivity [integral perturbed angular correlations (IPAC)] results [22,23] for ^{191,193}Ir. (b) Present and previous [7] IMPAC results for the Pt isotopes, with IPAC [24] and implantation-decay (I-D) IPAC [25] results. (c) Data for ^{162}Dy, from Ref. [26]. (d) IMPAC data for Nd and Sm [10] reevaluated with g factors from Refs. [27,28] and lifetimes from Ref. [18] and Nuclear Data Sheets.

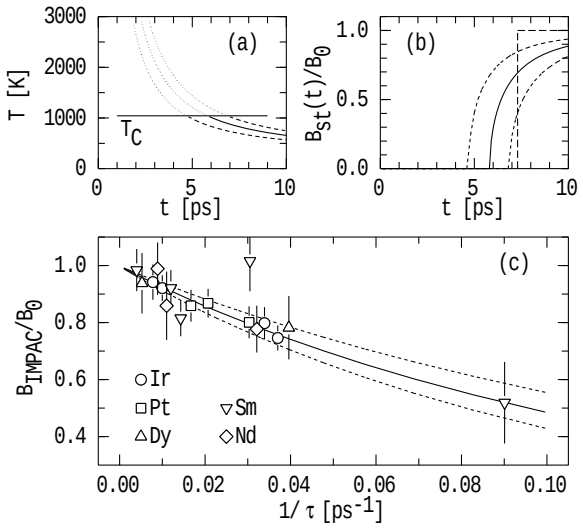

FIG. 3. Thermal-spike model interpretation of IMPAC data, Eqs. (3) and (4), for $T_1 = 8000 \pm 2000$ K. (a) Time dependence of the local temperature. (b) Time variation of the hyperfine field. The step function (long dashes) shows $B_{\rm st}$ implied Eq. (2) with $t_s = 7.3$ ps. (c) Resultant lifetime dependence of the IMPAC data. The solid line ($T_1 = 8000$) is not significantly different from that given by Eq. (2) with $t_s = 7.3$ ps.

thermal-spike lifetime of $t_s = 7.3$ ps, which falls within the expected range of between a few and ~10 ps [1,3].

The assumed time dependence of the hyperfine field embodied in Eq. (2) is clearly an oversimplification. We have therefore performed a more realistic modeling of the preequilibrium behavior of the hyperfine fields. Guided by the molecular dynamics calculations and discussion of Hsieh *et al.* [29], we assume that the local temperature, T, varies with time, t, approximately as

$$T(t) = T_1/t^q + T_0\,, \qquad (3)$$

where T_1 and q are parameters, and $T_0 = 300$ K is the ambient temperature. Noting that Eq. (3) has no physical implications until the local temperature falls below $T_{\rm C} = 1043$ K, the Curie temperature of iron, we put $q = 1.35$ [29] and treat T_1 as a fit parameter. We then assume that the hyperfine field follows the temperature dependence of the host magnetization, which for Fe is given approximately by the Brillouin function for $J = 1/2$. It follows that $B_{\rm st}(T)/B_0 = M(T)/M(0) = m(T)$, where $m(T)$ is the magnetization at temperature T relative to that at absolute zero, and $m(T) = \tanh[m(T)\,(T_{\rm C}/T)]$. The lifetime dependence of the observed hyperfine field is then given by

$$B_{\rm IMPAC}(\tau)/B_0 = \int_0^\infty m(T[t])e^{-t/\tau}dt/\tau\,. \qquad (4)$$

Fits to the IMPAC data obtained by numerically evaluating Eqs. (3) and (4) are shown in Fig. 3(c); the associated time dependencies of the local temperature and the hyperfine field are shown in Figs. 3(a) and 3(b). Since the solid-line fit to the IMPAC data in Fig. 3(c) is almost indistinguishable from Eq. (2) with $t_s = 7.3$ ps, Eq. (2) can be used as a convenient means of analyzing IMPAC data, provided it is kept in mind that t_s may then overestimate the time for which the hyperfine field is actually absent.

The behavior of the local temperature implied by the IMPAC data is similar to that found in molecular dynamics calculations. For example, 5 keV cascades in Cu take about 5 ps to fall to 1000 K [29], while the atomic rearrangements are over within 5 to 7 ps following 5 keV cascades in Si [6]. Bearing in mind that the energy deposited in collision cascades is several keV, even for MeV ions [1], it is reasonable to find that ~8 MeV ions with $140 < A < 200$ implanted into Fe give rise to local heating that takes about 6 ps after implantation to cool below 1000 K.

Our work can be compared with the in-beam Mössbauer measurements of Hardy *et al.* [30] and Chien *et al.* [31] on rare-earth ions implanted into rare-earth oxides following Coulomb excitation, in which the recoilless fraction was found to decrease as the lifetime of the probe state decreased. In those measurements the hyperfine fields could not be measured and the analysis reported in Refs. [30,31] is strongly dependent on a simplified thermal model. If the absence of the Mössbauer effect is taken as indicating violent atomic motion, these measurements suggest a thermal-spike lifetime in the

range between about 20 and 100 ps [2]. Although this is somewhat longer than we observe, better agreement is likely to emerge from a more rigorous analysis. Our IMPAC results support the thermal-spike interpretation of these Mössbauer data.

While the present work was in progress, Alfter *et al.* [26] reported evidence for lifetime-dependent hyperfine fields following the implantation of ^{162}Dy and ^{166}Er into iron. As shown in Fig. 2, their results for states in ^{162}Dy are consistent with an analysis based on Eq. (2) with $t_s = 7.3$ ps. However, as their IMPAC fields for ^{166}Er are about half of the strength expected, Alfter *et al.* propose a rather long relaxation process, of the order of 450 ps for Er in Fe, which they attribute to hindered exchange within the impurity-host spin system after implantation. Although other interpretations of their data are possible, this is an interesting suggestion that should be investigated further. Clearly, the time the impurity-host spin system takes to reach equilibrium after implantation could influence the interpretation of our data. At present, however, the data strongly suggest that the thermal-spike mechanism is dominant in the time up to about 10 ps.

In conclusion, we have observed picosecond-duration preequilibrium effects in hyperfine magnetic fields following ion implantation, consistent with the hyperfine field being absent for several picoseconds after implantation. The data can be interpreted in terms of a thermal-spike induced quenching of the hyperfine field and, as such, give the first direct measurement of the thermal-spike lifetime. In the future it will be possible to study aspects such as the influence of the electron-phonon coupling strength on the lifetime of the thermal spike [32] through measurements in different ferromagnetic hosts.

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[21] The observed lifetime dependence of the IMPAC fields cannot be attributed to electric quadrupole interactions caused by local damage, or to the possibility that the hyperfine fields might not be quite parallel to the applied field, because these effects imply *larger* effective fields for the shorter-lived states. Furthermore, because the lifetimes of the probe states in the present study are short ($<$130 ps), (i) electric quadrupole interactions are negligibly small and (ii) in the absence of any preequilibrium effects, the states experience the same average magnetic interaction whether there is a unique site or several sites, and whether or not the internal field is exactly parallel to the external field [9,13].
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